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Environmental Studies on Radioecological Sensitivity and Variability with Special Emphasis on the Fallout Nuclides ^{90}Sr and ^{137}Cs

Asker Aarkrog

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June 1979

RISØ-R-437

ENVIRONMENTAL STUDIES ON RADIOECOLOGICAL SENSITIVITY AND VARIABILITY WITH SPECIAL EMPHASIS ON THE FALLOUT NUCLIDES ^{90}Sr and ^{137}Cs

Part two

Appendices

by

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APPENDIX A

SAMPLING, ANALYSIS, MEASUREMENT AND ERROR

This appendix comprises summaries of sampling methods and analytical procedures including counting and an assessment of the lower limit of detection. The appendix is concluded by an estimate of the precision and accuracy of the data.

A.1. Sampling methods

To estimate a variable in a population (cf.B.2.), it is usually impossible to measure the whole population. Instead, a sample must be taken, i.e. a number of individuals are selected from the population and measured. This selection should be made so as to obtain a truly representative sample of the population, which is best done by random selection. In the present study, the various sample locations and sampling intervals were selected beforehand with a view to being representative of the area studied with respect to food production, rainfall and soil characteristics, and to ensure that seasonal and yearly variations were taken into account. Once these locations and sampling intervals had been determined, sampling was carried out randomly.

The present sampling programme was designed for a study of the environmental radioactive contamination from global fallout in Denmark, the Faroes and Greenland. As both population and food production were concentrated in Denmark ($5 \cdot 10^6$ inhabitants), most samples were collected there. On the other hand, Greenland ($5 \cdot 10^4$ inhabitants) and the Faroes ($4 \cdot 10^4$ inhabitants) were not neglected because the special environmental conditions and dietary habits in these territories can enhance the dietary intakes of radionuclides by the local populations; in other words, the radioecological sensitivity of humans may

be higher in the Faroes and Greenland than in Denmark. Sampling in the Faroes and Greenland was carried out with the help of doctors, pharmacists, and staff at telestations and meteorological stations. However, it was not always possible to obtain all samples specified in the programmes because communications, especially with remote locations in Greenland, were poor.

The Danish sampling programme was intended to yield data on the radioactive contamination of the various foods both where they were produced and at the site of consumption. The production levels were estimated from two main sampling systems: the seven dried milk factories (Fig.A.1.3.2), which yielded an estimate of the radionuclide levels in Danish milk products, and the eleven state experimental farms (Fig.A.1.1.3.1 and table A.1.1.6.2) from where samples of precipitation, soil, grain, potatoes, milk and fodder were obtained. The sampling system for the consumption site consisted of the 48 provincial towns ($1.14 \cdot 10^6$ inhabitants) in the 8 zones (Figs. A.1.4.2.1 and A.1.4.2.2) and Copenhagen ($1.35 \cdot 10^6$ inhabitants). The samples from this system were purchased in local stores and consisted of all diet components, besides simultaneous collection of individual samples of bread, dairy milk and vegetables.

The sampling methods that were applied in the present study are summarized below in the tables denoted A.1. followed by two digits identical to the numbering in the main text of the sample category in question. The sampling locations are shown in corresponding figures. More detailed information on sampling methods appears in the Risø Reports referred to in the table heads of the various samples. Sampling principles for assessment of radioactive contamination in the environment have, moreover, been dealt with in several publications (Har172, Wo66, Na76).

Table A.1.1.2. Sampling of air

	Small sampler (Ile 56-57)	Large sampler (RRD 60)
Method	25 ø cm paper filters	2 rectangular glass fibre filters, 56 x 48 cm^2 each
Size	300-400 m^3 day^{-1}	36000 m^{-3} day^{-1}
Locality	Risø 3 m above the ground	Risø 3 m above the ground
Period	Since 1956	Since 1960
Frequency	Continuous operation daily samples	Continuous operation half-weekly samples
Nuclides	^{90}Sr (^{89}Sr)	^{90}Sr (^{89}Sr), γ-emitters especially ^{137}Cs
Pooling before analysis	The daily samples for 1 month are combined	Radiostrontium on monthly aliquots. γ on half-weekly total samples
Remarks	Filter efficiency ~ 100%	Filter efficiency ~ 100%

Table A.1.1.3.1. Sampling of precipitation in Denmark

	Small rain bottles (Risø R 1) (RRD 60)	Ion exchange columns (Risø I 2) (RRD 61, RRD 62)	1 m^2 sampler (Risø R 3) (He 56-57)	Meteorology tower (Risø M 4) (RRD 59)	State exp. farms (SEFF) (RRD 62)
Method	Eight polyethylene bottles with polyethylene funnels	Five ion exchange columns with polyethylene funnels Dowex 50 x 8 Dowex 2 x 8	A quadratic stainless steel sampler	Eight polyethylene bottles at different heights in the 123 m tower	Three polyethylene bottles at each station with 25 ø cm funnels
Size	Sampling area of the 8 bottles, 0.24 m^2	Sampling area of the 5 samplers 0.33 m^2 (0.25 m^2 before 1970)	1 m^2	Each bottle had a funnel with a sampling area of 154 cm^2	Sampling area of the 3 bottles: 0.15 m^2
Locality	5 locations at Risø and 3 in the neighbourhood	5 locations at Risø identical to those of the rainbottles	Risø	Meteorology tower at Risø	(fig.A.1.1.3.1.) The state exp. farms
Period	Since 1960	Since 1962	1965-1970	1959-1974	Since 1962
Frequency	Continuous sampling; bottles exchanged monthly	Continuous sampling; columns exchanged monthly	Continuous sampling; water collected after rainfall	Continuous sampling; bottles exchanged monthly	Continuous sampling; bottles exchanged monthly
Nuclides	^{90}Sr (^{89}Sr)	^{90}Sr (^{89}Sr), in a few cases also γ-spectroscopy	^{90}Sr (^{89}Sr)	^{90}Sr (^{89}Sr), occasionally ^{137}Cs, ^{144}Ce	^{90}Sr (^{89}Sr)
Pooling before analysis	The eight bottles combined to one monthly sample	The five columns combined to one monthly sample	The daily samples combined to a monthly sample	The content of each bottle is analyzed	Bimonthly samples are analyzed from each station JAN-FEB, MAR-APR, etc.
Remarks	Since 1973 quarterly samples	Since 1973 quarterly samples		1971-1972 quarterly, in 1973 half-yearly and in 1974 yearly samples	Ledreborg was included from 1965

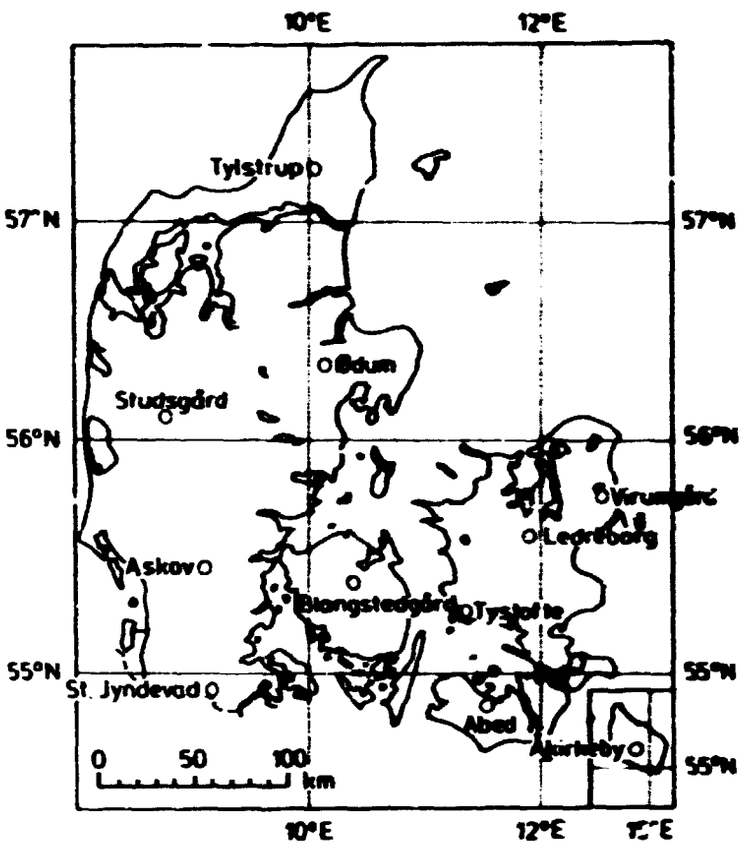

Fig. A.1.1.3.1. State experimental farms in Denmark. Sampling locations for precipitation (Table A.1.1.3.1.), soil (Table A.1.1.6.1.), grain (Table A.1.2.2.), grass and other fodder products (Table A.1.2.4.), potatoes (Table A.1.2.5.1.), and whole milk (Table A.1.3.2.); (cf. also table A.1.1.6.2.).

Table A.1.1.3.2. Sampling of precipitation in the Faroes and Greenland

	Meteorological rain gauge The Faroes (RRF 62)	Meteorological rain gauge Greenland (RRG 62)
Method	Standard rain gauge as used by Meteorological Institute	Standard rain gauge as used by Meteorological Institute
Size	200 cm^2 at each location	200 cm^2 at each location
Locality	Høyvik (Thorshavn) and Klaksvik (Fig. A.1.1.3.2.)	Upernavik, Godhavn Godthåb, Pr. Chr. Sund, Kap Tobin (Scoresbysund) (Fig. A.1.1.3.3.)
Period	Since 1962	Since 1962 (irregular)
Frequency	Continuous sampling. Precipitation samples are sent monthly to Denmark in polyethylene bottles	Continuous sampling. The samples are combined in polyethylene bottles and sent to Risø every quarter
Nuclides	^{90}Sr (^{89}Sr)	^{90}Sr (^{89}Sr)
Pooling before analysis	Monthly samples from each of the two stations are analyzed	Quarterly samples from each of the five stations are analyzed
Remarks	Due to evaporation during the storage of the bottles in the Faroes and Greenland, the concentrations found in rain water may be overestimated, but the total deposition is uninfluenced.	

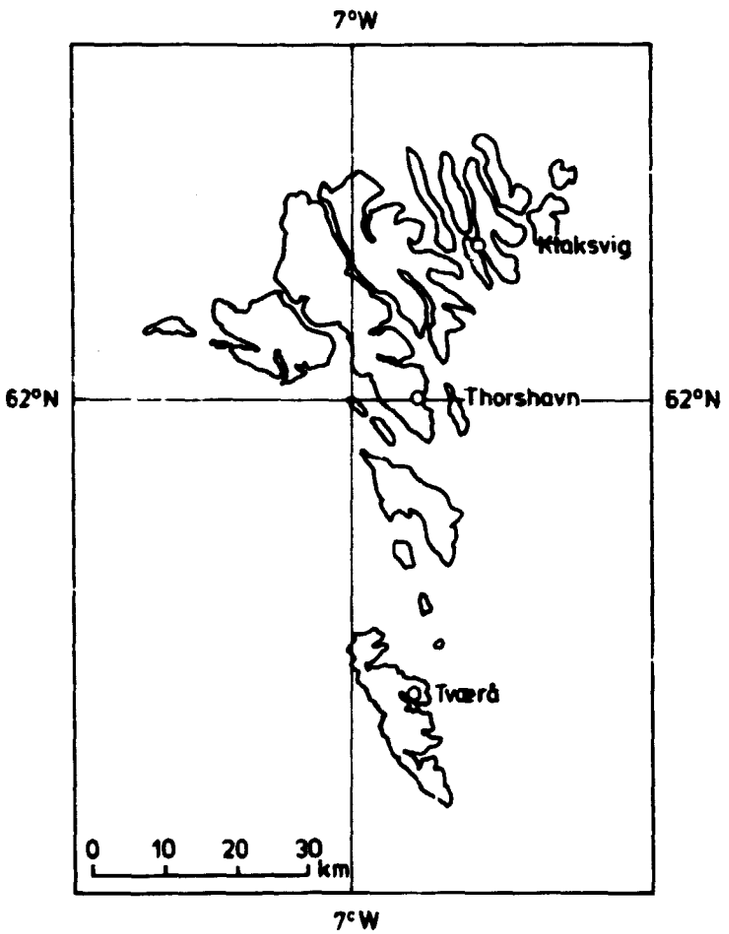

Fig. A.1.1.3.2. The Faroes. Sampling locations for precipitation (Table A.1.1.3.2.) drinking water (Table A.1.1.4.2.), sea water (Table A.1.1.5.), soil (Table A.1.1.6.1.), bread (Table A.1.2.3.), grass (Table A.1.2.4.), potatoes (Table A.1.2.5.1.), sea plants (Table A.1.2.7.), milk (Table A.1.3.3.), mutton (Table A.1.3.4.2.), fish (Table A.1.3.5.), whale (Table A.1.3.6.1.), sea-birds and eggs (Table A.1.3.6.2.), human bone (Table A.1.4.3.), and deciduous teeth (Table A.1.4.4.).

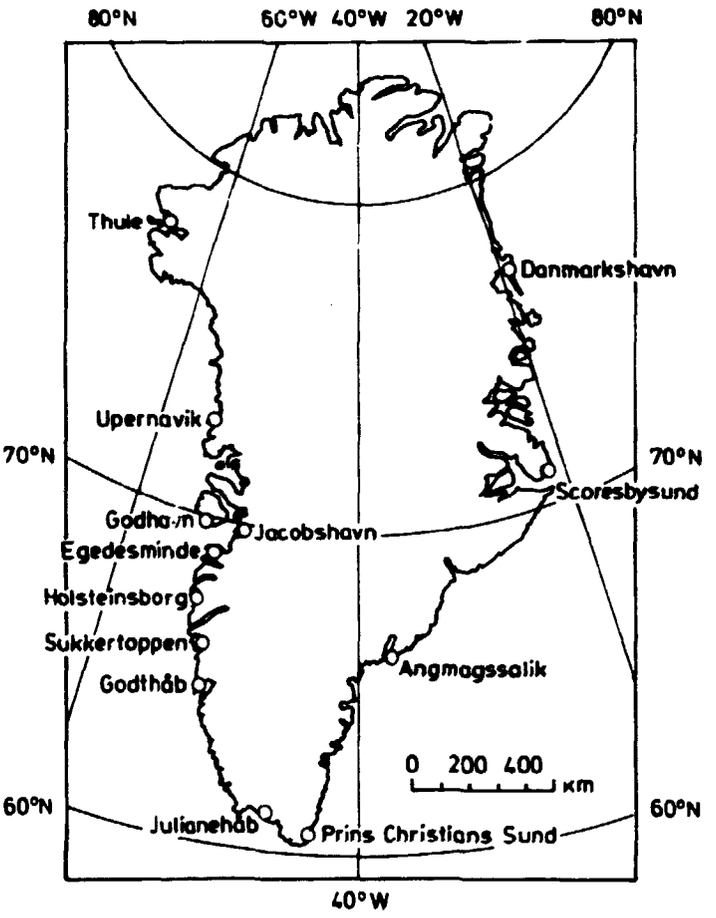

Fig. A.1.1.3.3. Greenland. Sampling locations for precipitation (Table A.1.1.3.2.), drinking water (Table A.1.1.4.2.), sea water (Table A.1.1.5.), soil (Table A.1.1.6.1.), grass (Table A.1.2.4.), lichen, moss and berries (Table A.1.2.6.), sea plants (Table A.1.2.7.), mutton, reindeer and musk ox (Table A.1.3.4.2.), fish (Table A.1.3.5.), whale and seal (Table A.1.3.6.1.), seabirds (Table A.1.3.6.2.), and deciduous teeth (Table A.1.4.4.).

Table A.1.1.4.1. Fresh water sampling in Denmark

	Ground water (RRD 61)	Stream water (RRD 71)	Lake water (RRD 71)	Drinking water (RRD 65)
Method	The water was pumped up from borings selected by the Geological Survey of Denmark	The water was collected directly in the polyethylene container from the surface	The water was collected from the surface in the same way a the stream water	The water was collected from taps in 10 l polyethylene bottles
Size	100 l in a polyethylene container	10 l	10 l	10 l from each town
Locality	Countrywide from 9 locations (Fig. A.1.1.4. 1.)	One stream in each of the eight zones (Fig. A.1.1.4. 2.)	One lake in each of the eight zones (Fig. A.1.1.4. 2.)	48 towns and Copenhagen Fig. A.1.4.2.1. and Fig. A.1.4.2.2.
Period	Since 1961	Since 1971	Since 1971	1965-1973
Frequency	Once a year in March	Every second year	Every second year	1965-68 in June and Dec. 1969-1973 in June only
Nuclides	^{90}Sr, Ca, Sr	^{90}Sr (^{137}Cs) Ca, Sr (K)	^{90}Sr (^{137}Cs) Ca, Sr (K)	^{90}Sr
Pooling before analysis	Individual samples analyzed	Individual samples analyzed	Individual samples analyzed	The samples from 6 towns were combined to a 60 l zone sample. Cph. was one sample
Remarks		Irregular sampling 1964-1970	Irregular sampling 1964-1970	

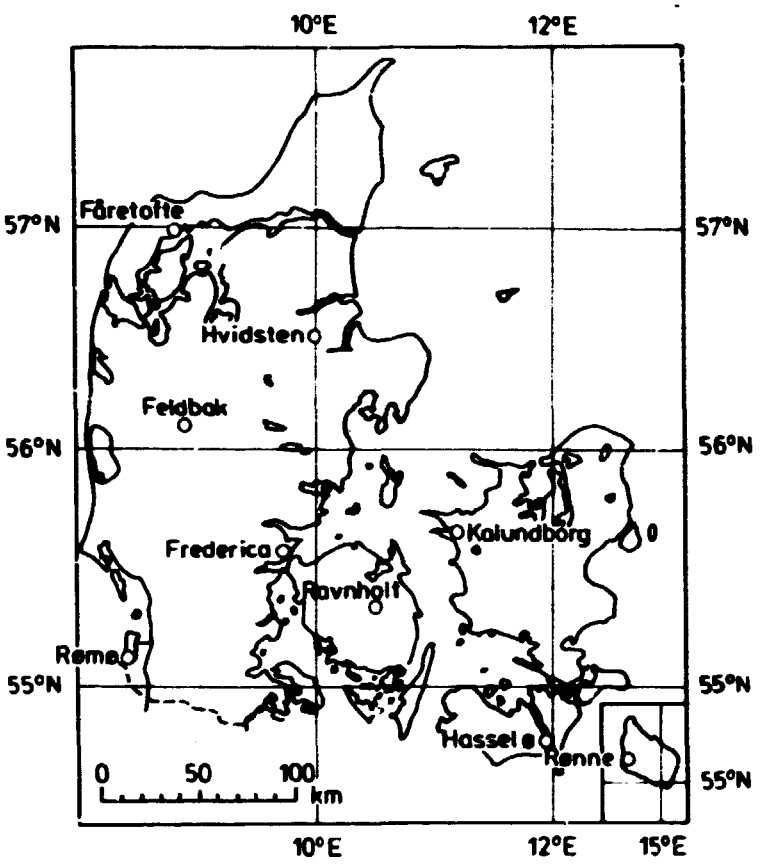

Fig. A.1.1.4.1. Ground-water sampling locations in Denmark (Table A.1.1.4.1.).

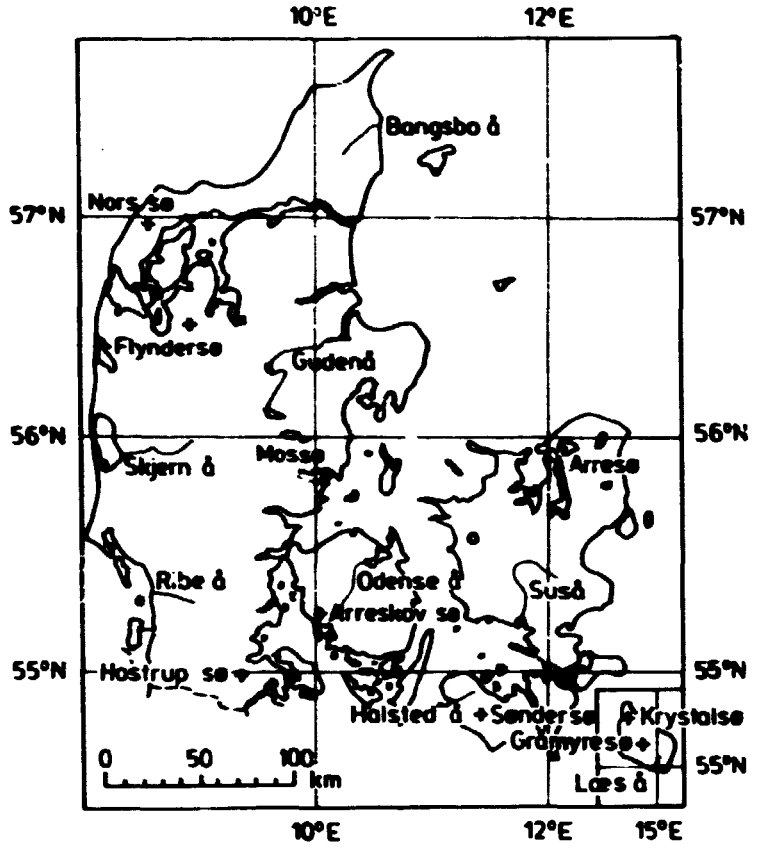

Fig. A.1.1.4.2. Sampling locations for fresh water from streams (å) and lakes (sø) (Table A.1.1.4.1.).

Table A.1.1.4.2. Drinking water sampling from the Faroes and Greenland

	Faroese drinking water (RRF 62)	Greenland drinking water (RRG 62)
Method	Tap water	Drinking water at the location
Size	10 l	10 l
Locality	Thorshavn, Klaksvig and Tvørå (Fig. A.1.1.3.2.)	Upernavik, Godhavn, Godthåb, Pr. Chr. Sund, Danmarkshavn. (Fig. A.1.1.4.3.)
Period	Since 1962	Since 1962
Frequency	Every second month	Every quarter (irregular)
Nuclides	^{90}Sr (^{89}Sr) (Ca)	^{90}Sr (^{89}Sr)
Pooling before analysis	Analysis of the individual samples	Analysis of the individual samples
Remarks	In 1967 supplementary samples were obtained from the water-works in Thorshavn	Other locations than the five mentioned above have occasionally also provided water samples

Table A.1.1.5. Sampling of sea water

	Inner Danish waters (RRD 62, RRD 70)	North Sea (RRD 72)	North Atlantic ocean (Faroes) (RRF 62)	Greenland waters (RRG 62)
Method	Pumping up through rubber tubing into a polyethylene container	Pumping up or using a 100 l water collector. Sampling performed by M/S DANA	DANA cruises and local people in the Faroes	DANA cruises and local people at telestations in Greenland
Size	50 l	50 l	50 l (occasionally 100 l)	50 l (occasionally 100 l)
Locality	Surface and bottom water from the locations shown in fig. A.1.1.5.	Generally surface waters from varying positions, cf.maps in Risø Reports (RRD 59-76)	Ocean water between Denmark and Iceland. Coastal waters at Thorshavn (generally surface water) (RRF 62-76)	Danmarkshavn, Pr. Chr. Sund, Godthåb Thule (RRG 62 - 76) surface water
Period	Since 1962	Since 1972	Since 1962	Since 1962
Frequency	In May-June and in Nov.-Dec.	Usually twice a year (Feb. and in summer)	Thorshavn 2-4 times per year. DANA cruises once a year (irregular)	Local samples in August. DANA cruises once a year (irregular)
Nuclides	^{90}Sr, ^{137}Cs (Ca) (Sr) Cl	^{90}Sr, ^{137}Cs Cl	^{90}Sr, ^{137}Cs Cl	^{90}Sr, ^{137}Cs Cl
Pooling	Neighbouring locations were pooled in the first years	No pooling but not all samples were analyzed for ^{90}Sr	No pooling	No pooling
Remarks	No analysis for ^{137}Cs until 1972. Recently, ^{90}Sr has not been determined in all samples		Deep water samples were occasionally obtained from the Atlantic Ocean	

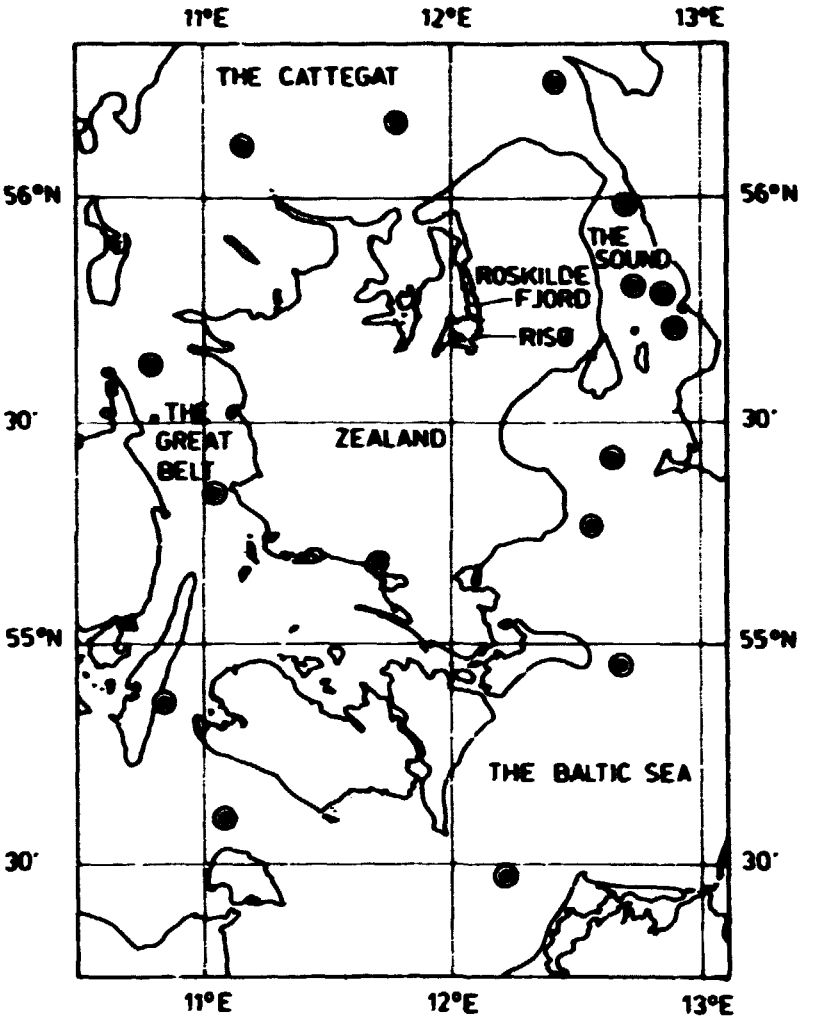

Fig. A.1.1.5. Sea water sampling locations around Zealand (Table A.1.1.5.).

Table A.1.1.6.1 - Soil and sediment sampling

	Uncultivated soil from state exp. farms (DDD 61)	Cultivated soil from state exp. farms (DDD 62, DDD 73, DDD 75)	Special soil samplings in Denmark	Soil from the Faroes (DDF 67) and Greenland	Sediments from Danish waters (DDD 75, DDD 76)
Method	Coring with a 60.5 mm (DDD 60) (later 65 mm) stainless steel tube in a 4 x 4 m^2 grid. The number of cores decreased with increasing sampling depth (DDD 59-76)		Coring with 65 mm tube. 13 cores in a 4 x 4 m^2 grid	Coring with 65 mm tube	Coring with a 135 mm stainless steel tube: HMPS (Ra 73)
Size	Sample weight: approx. 20 kg sampling depth: increased from 15 cm to 50 cm. Sample area: decreased from 945 cm^2 to 265 cm^2		Depth: 0-10, 10-20, 20-30 cm. Area: 630 cm^2.	50 cm depth in the Faroes and 35 cm in Greenland	Depth: 15-25 cm one core at each location
Locality	State experimental farms (Fig. A.1.1.3.1. and Table A.1.1.6.2.)		Ten locations (Fig. A.1.1.6.) with untouched soil (since 1950)	Thorshavn and Tvøroyri in the Faroes. Godthåb and Dundas (Thule Air Base) in Greenland	Roskilde Fjord The Sound Catregat. The Great Belt (Fig.A.1.1.5.)
Period	1961-1975 (except 1962)	1962, 1973 and 1975	1970 (cf. Fig. 1.6.2.)	Faroes: April 1967. Greenland: Aug. 1970	Since 1973
Frequency	Usually once a year in Sept. (cf. fig. 1.6.1.)		In 1970		Once or twice a year at the various locations
Nuclides	^{90}Sr, ^{137}Cs ($^{239,240}Pu$) (Cs) (Sr) (H)		^{90}Sr, ^{137}Cs	^{90}Sr, ^{137}Cs	^{137}Cs, $^{239,240}Pu$
Pooling before analysis	Usually an aliquot of 500 g of dried soil is analysed after thorough blending of the total sample. ^{137}Cs is determined in 2 kg		500 g for ^{90}Sr determination and 2 kg for ^{137}Cs measurement	(cf. D.1.6.1.3.)	Three cm thick slices are analysed down to full sample depth
Remarks	10 cm sections were analysed in some samplings to examine the vertical distribution of the activity (Fig. 1.6.2.)		Not published in Risø Reports		

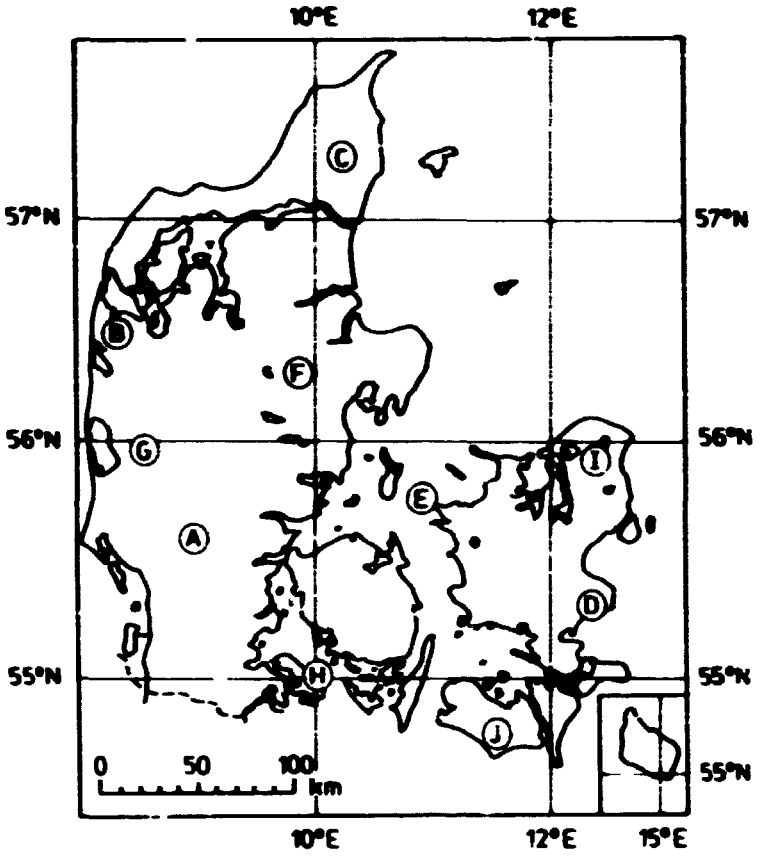

Fig. A.1.1.6. Soil sampling locations of special soil sampling in September 1970 (cf. Table A.1.1.6.1.). The annual mean amounts of precipitation (No 59-77) were: A: 800-850 mm; B: 700-750 mm; C: 600-650 mm; D: 500-550 mm and E: 400-450 mm. Locations F-J were all in the range 650-700 mm.

Table A.1.1.6.2. Characteristics of the sampling locations for soil, precipitation and various agricultural products in Denmark (cf. Fig. A.1.1.3.1.)

Location (Lat. N) (Long. E)	Annual mean precipitation in mm (Lu 71)	Texture class (Lu 71) (cultivated soil)	Uncultivated soil locations		
			Ferti-lizers	Cattle	Remarks
TYLstrup (57° 09') (10° 03')	661	loamy sand	no	no	meadow
[illegible]Um (56° 13') (10° 10')	658	sandy clay loam	yes	yes	meadow nearly surrounded by a stream
STUdegaard (56° 01') (8° 57')	833	loamy sand	no	no	heather
ASRov (55° 31') (9° 00')	848	sandy loam	yes	no	grass field
JYNdevad (54° 56') (9° 09')	824	sand	no	no	fallow field
BLAngstedgaard (55° 25') (10° 20')	611	sandy clay loam	no	no	fallow field
TYStofte (55° 16') (11° 20')	553	sandy loam	yes	no	grass field
VIRungaard (55° 44') (12° 35')	685	sandy loam	yes	yes	grass field
ABEd (54° 52') (11° 17')	615	sandy clay loam	yes	yes	low-lying meadow
AKIrkeby (55° 04') (14° 33')	641	sandy clay loam	yes	yes	grass field
RISø (55° 40') (12° 08')	517	sandy clay loam	no	yes	two sites 2 km apart, grass field
LEDreborg (55° 36') (12° 00')	was similar to RISø with respect to precipitation and soil characterization				

Table A.1.2.2. Sampling of grain in Denmark

	Annual samples of rye, barley, wheat and oats from the state experimental farms (RRD 59, RRD 61)	Special rye sampling 1959 (RRD 60)	Greenhouse experiment (RRD 62)
Method	Samples of grain from the harvest are collected by staff at the state experimental farms	Rye samples were collected by the staff at 27 Danish mills	Various grain species were grown inside and outside a greenhouse
Size	2 kg	1 - 2 kg	1 - 2 kg grain and 1 - 2 kg straw
Locality	The state experimental farms (Fig. A.1.1.3.1.)	19 locations in Jutland 4 in Funen 3 in Zealand 1 in Lolland	Risø
Period	Since 1959	In 1959	1962-1966
Frequency	Annually at harvest	-	At harvest
Nuclides	^{90}Sr, (^{89}Sr), (^{54}Mn), ^{137}Cs, ($^{239,240}Pu$), Ca, K, Sr	^{90}Sr, Ca	^{90}Sr, (^{89}Sr) (^{54}Mn), ^{137}Cs Ca, K
Pooling before analysis	Individual samples analyzed	Individual samples analyzed	Individual samples analyzed
Remarks	Rye: Secale cereale (spring and winter varieties) Barley: Hordeum sp. (spring and winter varieties) Wheat: Triticum sp. (spring and winter varieties) Oats: Avena sp. (spring varieties)	This sampling suggested that the state exp. farms represented the country average with regard to ^{90}Sr in rye	The air supply to the greenhouse was filtered in order to avoid direct contamination

Table A.1.2.3. Sampling of bread and other cereal products

	Danish bread (RRD 62)	Faroese bread (RRF 64)	Oat grits and rice (RRD 60)
Method	Samples of rye and white bread were purchased in local shops	Samples of Faroese rye and white bread were purchased in Thorshavn	Usually purchased in shops in Copenhagen
Size	1 rye loaf (100% extraction) and 2 - 3 white loaves (75% extraction) in each shop	1 rye loaf (~80% extraction) 2 - 3 white loaves (~75% extraction)	1 - 2 kg
Locality	6 towns in each of the 8 zones + Copenhagen (Fig. A.1.4.2.1. and Fig. A.1.4.2.2.). Since 1973 in 8 towns only + Copenhagen (Fig. A. 1.4.2.3).	Thorshavn (Fig. A.1.1.3.2.)	Copenhagen
Period	Irregularly 1960-1961 (RRD 60, RRD 61), since 1962 regularly	Since 1964	Since 1959, (irregularly)
Frequency	In June and December. Since 1971 in June only. During 1962 every quarter	In June and December	Once a year in December
Nuclides	^{90}Sr, ^{137}Cs (^{54}Mn) Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Samples of each type of bread are pooled from the towns within each zone, thus 9 rye loaf samples and 9 white loaves analyzed each time	Analysis of each type of bread from the individual samplings	Individual samples analyzed
Remarks	Other types of bread sample have also been investigated: crisp bread, light rye bread (80% extraction) and coarse white bread of rye (70% extraction)	A supplementary sampling took place in in April 1967 (RRF 67)	A special sampling of unpolished rice took place in 1970 (RRD 70)

Table A.1.2.4. Sampling of grass and other fodder products

	Risø grass (No 56-57)	Grass from state exp. farms (BBB 62)	Faroese grass (BBF 62)	Greenlandic grass (BBG 62)	Various cattle fodders BBD 64, BBD 74)
Method	1 m^2 sheared to 2-3 cm above ground	1 m^2 sheared to from comp' pasture (cf. Table A.1.3.3.1)	Sheared to grazing height, carried out by local people	Sheared by local people	Hays, silage and concentrates collected by staff at state exp. farms
Size	½ - 1 kg fresh weight (~ 1 m^2)	~ 1 kg fresh weight (~ 1 m^2)	~ 1 kg fresh weight	~ 1 kg fresh weight	~ 1 - 2 kg
Locality	At 33 locations up to a distance of 16 km from Risø (Fig. A.1.2.4.)	At the state exp. farms keeping cows, i.e. all localities except Vi-ungård (Fig. A.1.1.3.1.)	Thorshavn (Fig. A.1.1.3.2)	Usually West Greenland, Exp. farmlands, Holsteinsborg, Sukkertoppen, Godthåb, Julianehåb (Fig. A. 1.1.3.3.)	Askov, Studsgård, Ledreborg, Odum, Tystofte, Jyndevad (Fig. A.1.1.3.1)
Period	Since 1957	1962-1973 and 1977	Since 1962	Since 1962 (Irregularly)	Since 1963 (irregularly)
Frequency	Quarterly (but more frequently earlier)	In Sept (once a year)	Usually twice in the grazing season	Usually once during the summer (July-Aug.)	Once a year
Nuclides	^{90}Sr (^{89}Sr) Ca	^{90}Sr (^{89}Sr) (^{131}I) ^{137}Cs K, Ca	^{90}Sr (^{89}Sr), ^{137}Cs K, Ca	^{90}Sr, (^{89}Sr), ^{137}Cs K, Ca	^{90}Sr, ^{137}Cs K, Ca
Pooling before analysis	All samples collected in a quarter are pooled before ^{90}Sr analysis	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples
Remarks		Grass from June 1962-1964 in June also		Samples often more like hay than grass	

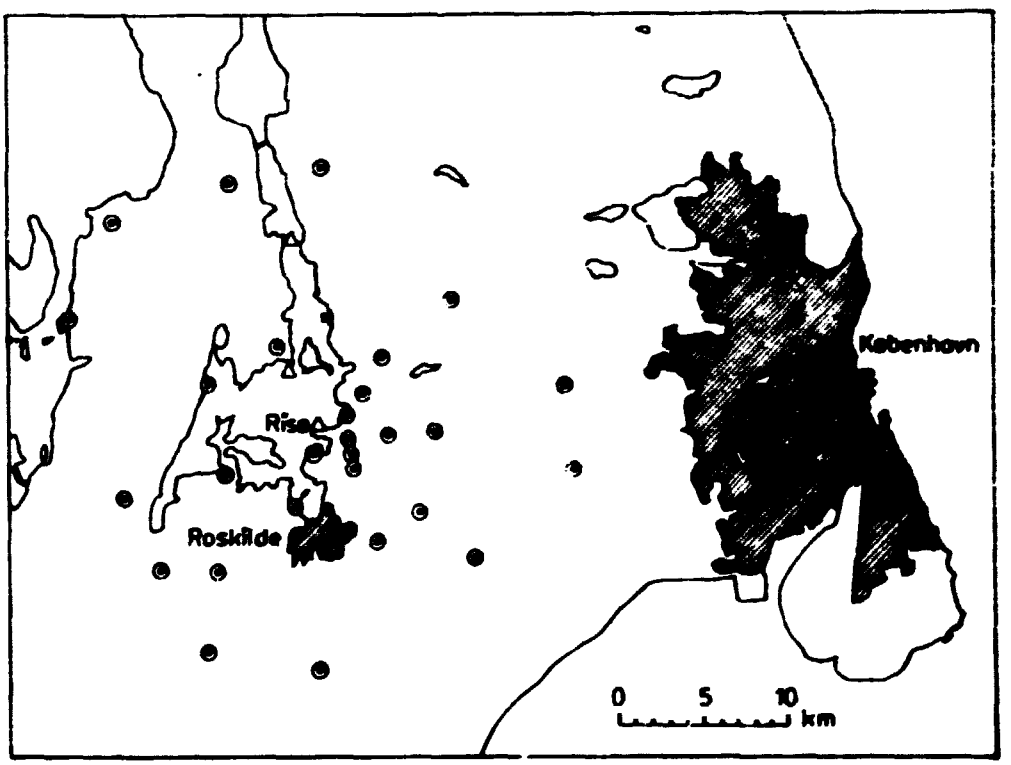

Fig. A.1.2.4. Sampling locations around Risø for grass (● Table A.1.2.4.) and sea plants (△ Table A.1.2.7.).

Table A.1.2.5.1. Sampling of cabbage, carrots, potatoes and apples

	Danish cabbage (RRD 61, RRD 64, RRD 66)	Danish carrots (RRD 61, RRD 64 RRD 66)	Danish potatoes (RRD 60)	Faroese potatoes (RRF 62)	Danish apples (RRD 64, RRD 66)
Method	Purchased in markets or at local dealers	Purchased in markets or at local dealers	The samples are collected by staff at the state exp. farms	The samples are collected by local people	Purchased from local dealers
Size	Approx. 2-4 kg from each dealer	Approx. 2-4 kg from each dealer	~ 5 kg	~ 2 kg	2 - 4 kg
Locality	The 8 zones and Copenhagen along with the total diet sampling in Dec. (cf. table A.1.4.2.)	As for cabbage, until 1965. Since 1966, Fig. A.1.2.5.1.	The state exp. farms (Fig. A. 1.1.3.1.)	Usually in the the neighbourhood of Thorshavn (Fig. A.1. 1.3.2.)	The 8 zones (cf. carrots)
Period	Since 1961 (except 1962)	Since 1961 (except 1962)	Since 1959	Since 1962 (irregularly)	Since 1963
Frequency	Once a year (Dec.)	Once a year (Sept.-Dec.)	Once a year in Sept.-Oct.	Usually once a year (irregularly)	Once a year in Sept.-Oct.
Nuclides	^{90}Sr, ^{137}Cs Ca, (Sr), K	^{90}Sr, ^{137}Cs Ca, (Sr), K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	A and B towns were usually analysed separately, but towns within a zone were combined	Samples within a zone usually pooled	Individual samples analyzed	Individual samples analyzed	Samples pooled into zone samples
Remarks	In 1962 cabbage and carrots only represented by Blangstedgård (Table A.1.2.5.2.)				

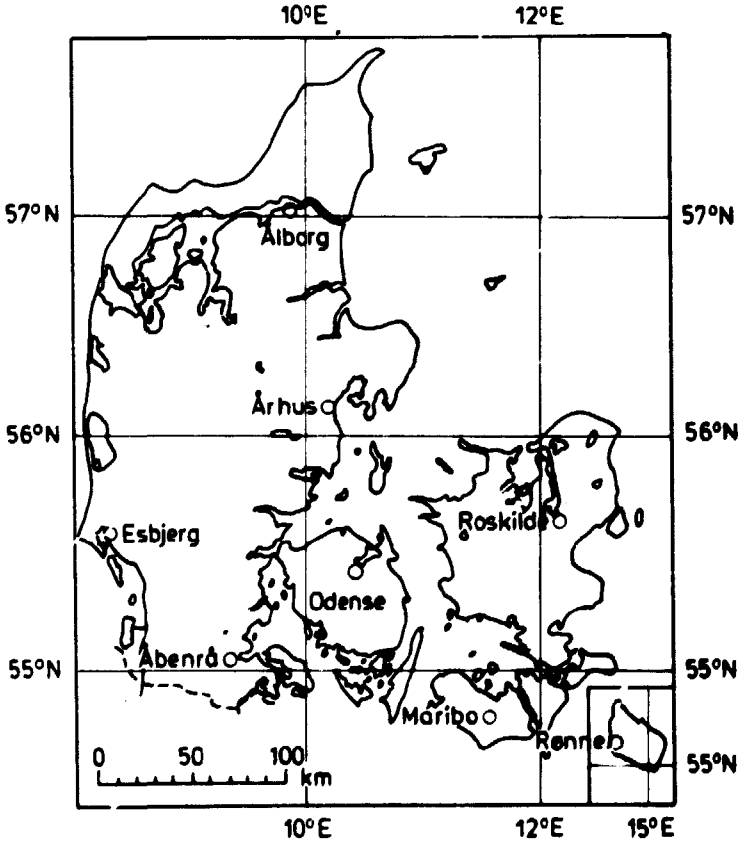

Fig. A.1.2.5.1. Sampling locations for vegetables and fruit since 1966. (Tables A.1.2.5.1. and A.1.2.5.2.).

Table A.1.2.5.2. Sampling of various vegetables and fruit

	Brussels sprouts sampling (RRD 62, RRD 63)	Roskilde 1960 sampling (RRD 60)	Blangstedgård sampling (RRD 62)	Danish countryside sampling (RRD 62, RRD 64, RRD 66)	Imported fruit, (RRD 63) coffee and tea (RRD 63)
Method	Leaves of brussels sprouts gathered throughout the growing season by local staff	All available vegetables and fruit purchased at the market	All available vegetables grown at Blangstedgård collected by the staff	Vegetables and fruit, apart for cabbage, carrots and apples, purchased in local shops	Samples purchased from supermarkets in Copenhagen
Size	Approx. 1 kg fresh weight	1 - 2 kg fresh weight	1 - 2 kg	1 - 2 kg	1 - 2 kg from each of 3 stores
Locality	Six exp. horticultural stations for vegetables (Fig. A.1.2.5. 2.)	Roskilde (Zealand)	Blangstedgård (Fig. A.1.2.5. 2.)	The 8 zones + Copenhagen (cf. carrots)	Copenhagen
Period	1962-1963	1960 (June-Dec.)	1962-1963	Since 1962, (irregularly)	Since 1962, (irregularly)
Frequency	Every month (in 1963 every fortnight)	Every week	Monthly or every fortnight	Once a year (Sept.-Dec.)	Usually in December
Nuclides	^{90}Sr, ^{89}Sr ^{137}Cs K, Ca, Sr	^{90}Sr Ca, Sr	^{90}Sr, ^{89}Sr ^{137}Cs K, Ca, Sr	^{90}Sr, ^{89}Sr, ^{137}Cs K, Ca, Sr	^{90}Sr, ^{137}Cs K, Ca, Sr
Pooling before analysis	Analysis of individual samples	Weekly samples pooled into monthly samples of each species	Analysis of individual samples	Analysis of zone samples	Samples from the 3 stores pooled before analysis
Remarks		These samplings provided the samples termed "leaf" and "root" vegetables in Figs. 2.5.1.3.-2.5.1.6. and 2.5.1.7.-2.5.1.10., respectively.			Tea and coffee are prepared as drinks before analysis

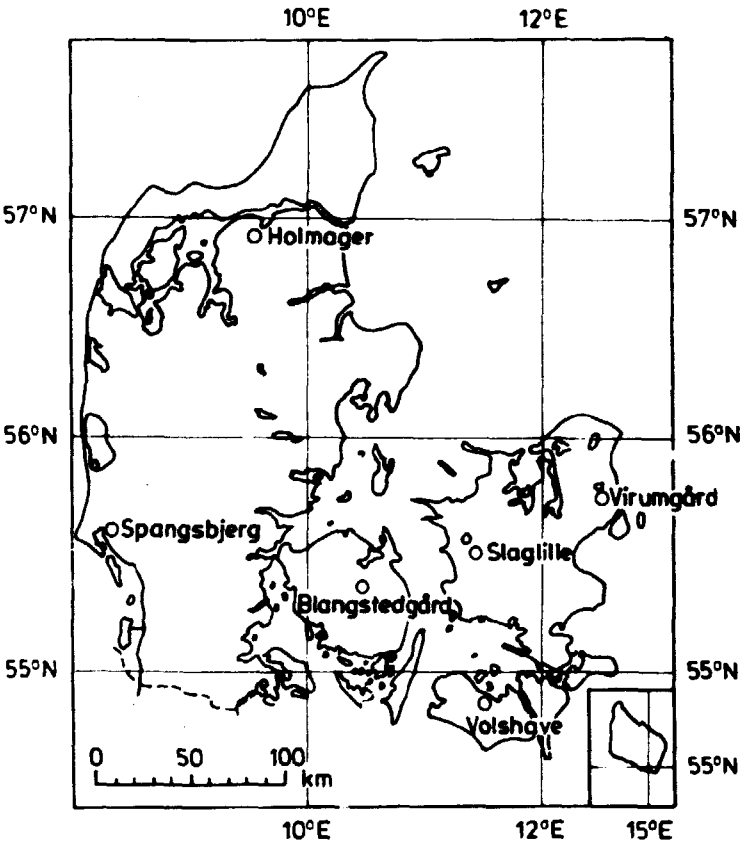

Fig. A.1.2.5.2. Sampling locations for brussels sprouts 1962-1963 (Table A.1.2.5.2.).

Table A.1.2.6. Sampling of Greenlandic lichen, moss and berries

	Lichen	Moss	Berries
Method	Picked by local people	Picked by local people	Collected by local people
Size	~ 1 kg (but often less)	~ 1 kg or less	~ 1 kg or less
Locality	Usually West Greenland (Egedesminde, Holsteinsborg, Sukkertoppen, Godthåb, Julianehåb) but also from other locations (Fig. A.1.1.3.3.)	Various locations in most parts of Greenland	Especially West Greenland
Period	Since 1962 (irregularly)	Since 1964 (irregularly)	Since 1962 (irregularly)
Frequency	Usually once during summer (July-Aug.)	Summer (July-Aug.)	Summer (July-Aug.)
Nuclides	^{90}Sr, (^{89}Sr), ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples
Remarks	These samples were not well defined as regards species, parts of the plant and weight by unit area		Crowberries and blueberries

Table A.1.2.7. Sampling of sea plants

	Roskilde Fjord (RRD 59)	The Sound (RRD 61, Pet 62)	Faroese waters (RRD 62)	Greenlandic waters (RRG 62)
Method	Collecting from boat by rake	Collecting from boat by rake (Danish Navy)	Samples collected by local people together with sea water samples	Samples collected by local people together with sea water samples
Size	~ 2 kg wet weight	~ 2 kg wet weight	1 - 2 kg	1 - 2 kg
Locality	3 locations in Roskilde Fjord (Fig. A.1.2.4.)	The Sound (Fig. A.1.1.5.)	Off Thorshavn (Fig. A.1.1.3.2.)	Along the Greenland coast (Fig. A.1.1.3.3.)
Period	Since 1959	1961 (March-Dec.)	Since 1962	Since 1962 (irregularly)
Frequency	Approx. once every quarter	Approx. twice during the period	Usually in April and August	Usually once in the summer (July-August)
Nuclides	^{90}Sr (^{137}Cs) Ca (Sr)	^{90}Sr Ca, Sr	^{90}Sr (^{137}Cs) Ca (Sr)	^{90}Sr (^{137}Cs) Ca (Sr)
Pooling before analysis	Samples pooled from the 3 locations to make half-yearly samples	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples
Remarks	The species were were: Fucus vesicolosus and Zostera marina	Five species	Laminaria and Fucus species	Laminaria and Fucus species

Table A.1.3.2. Sampling of Danish milk and milk products

	Dried milk (RRD 59, RRD 61)	Fresh dairy milk (RRD 61, RRD 62, RRD 63)	Untreated whole milk exp. farm (RRD 62)	Untreated whole milk (RRD 59)	Cheese (RRD 63)
Method	Dairy inspectors collected samples	Purchased in local dairies	Collected directly at the state exp. farms together with grass (cf. table A.1.2.4.)	Collected at a farm near Risø	Purchased in Copenhagen
Size	1 - 2 kg at each factory	2 l in each dairy	2 - 4 l at each farm	1 - 2 l each time	~ 1 kg
Locality	The seven dried milk factories shown in Fig. A.1.3.2.	In the 8 zones + Copenhagen (Fig. A.1.4.2.1., Fig. A.1.4.2.2. and Fig. A.1.4. 2.3.)	At the state exp. farms (Fig. A.1.2.3.1.)	Svaleholm until 1968, later other neighbouring farms	Copenhagen
Period	Since 1959	Since 1961	1962-1970 and in 1977	Since 1959	Since 1963 (irregularly)
Frequency	Monthly samples	June and December	In September	Half-weekly samples	Once a year
Nuclides	^{90}Sr, (^{89}Sr) (^{131}I) ^{137}Cs K, Ca	^{90}Sr, (^{89}Sr) (^{131}I) ^{137}Cs K, Ca	^{90}Sr, (^{89}Sr) (^{131}I) ^{137}Cs K, Ca	^{90}Sr, (^{89}Sr) (^{131}I) ^{137}Cs K, Ca	^{90}Sr, ^{137}Cs K, Ca
Pooling before analysis	One sample from each month and location	Samples pooled into zone samples before analysis	Analysis of individual samples	Analysis of monthly pooled samples; since 1965 quarterly pooled samples	Analysis of individual samples
Remarks	Only four factories until Sept. 1961. Kalundborg was replaced by Ringsted in 1969 (RRD 69)	In 1962-1964 samples were also collected in March and Sept.	In 1963-1964 also milk from June		

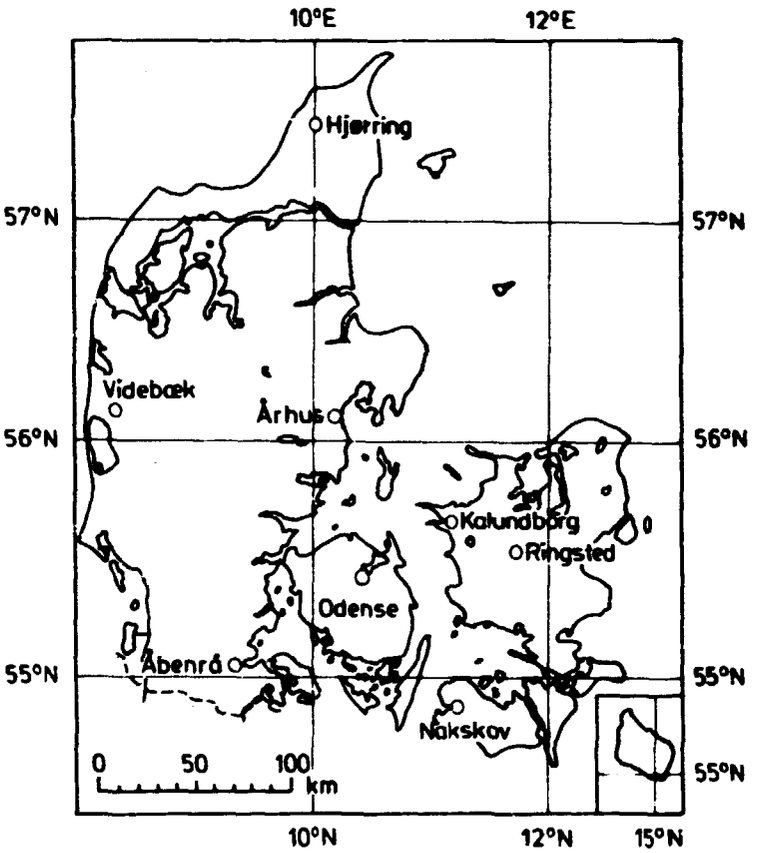

Fig. A.1.3.2. Sampling locations for dried milk (Table A.1.3.2.). In 1959-1961 the samples were taken at Hjørring, Åbenrå, Odense and Nakskov. In 1962-1968 samples were furthermore obtained from Videbæk, Århus and Kalundborg. Kalundborg was replaced by Ringsted in 1969.

Table A.1.3.3. Sampling of Faroese milk

	Fresh dairy milk (RRF 62)	Other milk products (RRF 63)
Method	Samples collected at local dairies and sent to Thorshavn (Tjaldur Apotek)	Butter and cheese occasionally obtained from dairies
Size	1 l milk from each dairy each week	~ 1 kg
Locality	Thorshavn, Klaksvik, Tvorå (Fig. A.1.1.3.2.)	Thorshavn (usually)
Period	Since 1962	Since 1963 (irregularly)
Frequency	Weekly sampling	Once a year
Nuclides	^{90}Sr, (^{89}Sr), (^{131}I), ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs, Ca, K
Pooling before analysis	Pooled into monthly samples from each location	No pooling
Remarks	Samples are preserved with formaline	

Table A.1.3.4.1. Sampling of Danish meat

	Beef and pork from Copenhagen (RRD 62, RRD 63)	Veal from Copenhagen (RRD 63)	Countrywide pork and beef sampling (RRD 63, RRD 64)
Method	The samples are purchased in three big stores in Copenhagen	Purchased in three big stores together with beef	Purchased at local dealers
Size	Approx. 1 kg of each kind of meat from each store	~ 1 kg	~ 0.25 kg in each shop
Locality	Copenhagen	Copenhagen	The 8 zones (Fig. A.1.4.2.1. and Fig. A.1.4.2.2.)
Period	Since 1962	1963-1970	1963-1964
Frequency	In March, June, September and December. Until 1966 only 1-3 samplings every year	As for beef and pork	Dec. 1963, June 1964 Dec. 1964
Nuclides	(^{90}Sr), ^{137}Cs, (Ca), K	^{90}Sr, ^{137}Cs, Ca, K	(^{90}Sr), ^{137}Cs, (Ca), K
Pooling before analysis	Each kind of meat is pooled from the 3 stores before analysis	As for beef and pork	In 1963: zone samples. In 1964: country samples (the zones were pooled)

Table A.1.3.4.2. Sampling of Faroese and Greenlandic meat

	Faroese lamb and mutton (RBF 62)	Greenlandic lamb and mutton (RBG 62, RBD 61)	Greenland reindeer (RBD 61, RBG 62)	Greenland musk ox (RBG 64)
Method	Collected by local people	From local people, or purchased from Royal Greenland Trade Company	From local people or purchased from Royal Greenland Trade Company	From local hunters
Size	1 - 2 kg	1 - 2 kg	1 - 2 kg	1 - 2 kg
Locality	Usually Thorshavn (Fig. A.1.1.3.2.)	S.W. Greenland (Julianehåb) (Fig. A.1.1.3.3.)	West Greenland between Egedesminde and Julianehåb	East Greenland between Danmarkshavn and Scoresby Sund (Fig. A.1.1.3.3.)
Period	Since 1962	Since 1961 (irregularly)	Since 1961	Since 1964 (irregularly)
Frequency	Usually twice a year	Once a year	Usually twice a year; late winter and autumn	Once a year (winter)
Nuclides	^{90}Sr, ^{137}Cs Ca, K	(^{90}Sr), ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples	Analysis of individual samples
Remarks	Also bone analysis	Also bone analysis	Also bone analysis	Also bone analysis

Table A.1.3.5. Sampling of fish

	Danish fish (RRD 61, Pet 62)	Faroese fish (RRF 63)	Greenlandic fish (RRG 62)
Method	Samples are purchased from fish dealers in Zealand, or fished out when sea-water sampling (Table A.1.1.5.)	Samples of cod and haddock fillets are purchased in Thorshavn	The samples are obtained through local people or purchased from the Royal Greenland Trade Company
Size	2 - 4 kg	2 - 4 kg	2 - 4 kg
Locality	Inner Danish waters: The Sound, Cattegat and the Great Belt. In 1970 from the North Sea (RRD 70)	North Atlantic	Greenlandic coastal waters (mostly West Greenland)
Period	Since 1961	Since 1963	Since 1962
Frequency	Usually once a year (in autumn or early winter)	4 - 6 times a year	Once a year, usually summer
Nuclides	^{90}Sr, ^{137}Cs Ca (Sr), K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Individual fish of same species are normally pooled if weighing less than 1 kg	The individual species are analyzed from each sampling	As for Danish fish
Remarks	The species are: cod, plaice, herring and occasionally eel, salmon, mackerel, garfish and trout	These samples were usually free of bone	Cod, Greenland halibut, salmon, trout and angmagsatter

Table A.1.3.6.1. Sampling of sea mammals

	Faroese whale (RRF 62)	Greenland whale (RRG 62)	Greenland seal (RRG 62)
Method	From local people	From local people or through Royal Greenland Trade Company	From local people or through Royal Greenland Trade Company
Size	2 - 4 kg (meat) Grind whale	2 - 4 kg (meat) Piked whale mostly	2 - 4 kg (meat) Various arctic species
Locality	Faroese waters	Greenlandic waters	Greenlandic waters
Period	Since 1962 (irregularly)	Since 1962 (irregularly)	Since 1962 (irregularly)
Frequency	Once a year	Once a year	Once a year
Nuclides	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Individual samples analyzed	Individual samples analyzed	Individual samples analyzed
Remarks			Also bone analysis

Table A.1.3.6.2. Sampling of sea-birds and hens' eggs

	Faroese sea-birds (RRF 62)	Greenlandic sea-birds (RRG 62)	Danish hens' eggs (RRD 63)	Faroese hens' eggs (RRF 63)
Method	From local people	From local people	Purchased in three large stores in Copennagen	Sampled at local farms near Thorshavn
Size	2 - 4 kg	2 - 4 kg	2 - 4 kg	2 - 4 kg
Locality	Faroes	West Greenland mostly	Copenhagen (produced countrywide)	Faroes
Period	Since 1962 (irregularly)	Since 1962 (irregularly)	Since 1963	Since 1963
Frequency	Once a year	Once a year	Once a year (usually in December)	Usually twice a year (summer and winter)
Nuclides	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K	^{90}Sr, ^{137}Cs Ca, K
Pooling before analysis	Individual species analyzed	Individual species analyzed	Eggs from each sampling pooled into one analysis	Eggs from each sampling pooled
Remarks	Fulmar Petrel Razorbill Puffin Guillemot	Eider Guillemot Occasionally eggs		

Table A.1.4.2. Total diet sampling

	Countrywide sampling from food stores ("Diet C") (RRD 61, RRD 63, RRD 73)	Local sampling of meals from private households (RRD 62) and canteens (RRC 63)
Method	The individual foodstuffs (milk, bread, vegetables, fruit, meat, fish, eggs, beer, coffee and tea) were purchased in local shops (in Copenhagen in super-markets)	Samples were collected as "doubles" of individual meals, or purchased in a canteen (Risø)
Locality	The 8 zones and Copenhagen: 1961-1962: Fig. A.1.4.2.1. 1963-1972: Fig. A.1.4.2.1. and Fig. A.1.4.2.2. Since 1973: Fig. A.1.4.2.3.	Zealand
Period	Since 1961	1960 (canteen) (RRD 60) 1962 (twice) (RRD 62) 1964 (RRD 64) 1965 (RRD 65) diet of donors of mother's milk (Table A.1.4.6.)
Frequency	In June and December	Usually once a year (in autumn) but in 1962 both in March and September
Nuclides	^{90}Sr, (^{89}Sr), (^{54}Mn), ^{137}Cs Ca, Sr, K	^{90}Sr, ^{137}Cs Ca, Sr, K
Pooling before analysis	Samples within a zone pooled (except 1963-1972) when two sets of towns (A and B) within each zone are analyzed (RRD 63)	Each day's ration was analyzed
Remarks	The samples are uncooked, but inedible parts (e.g. peel and bones) are removed before analysis	These samples were all prepared for eating

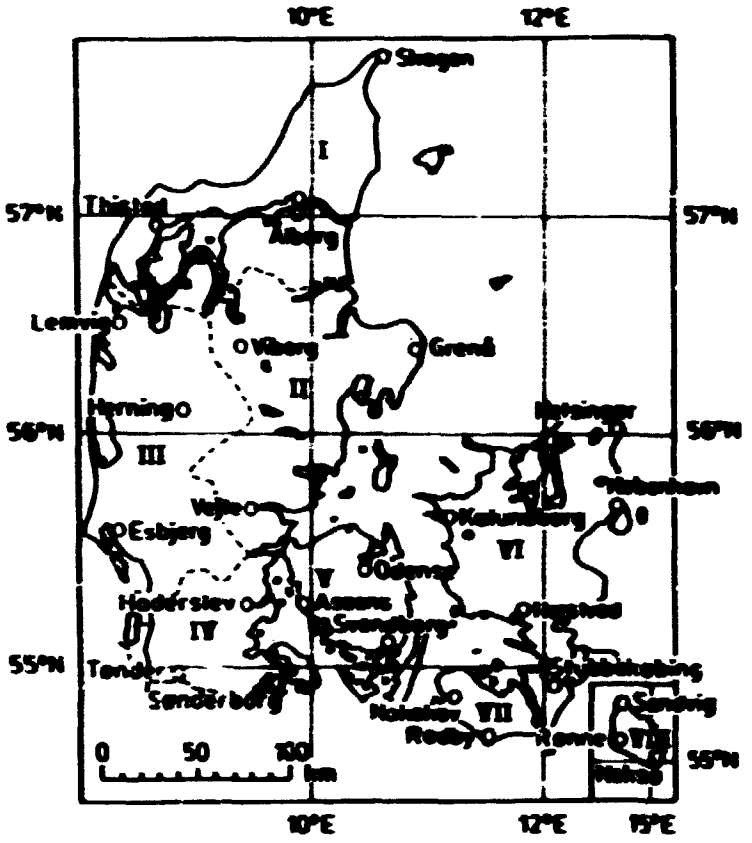

Fig. A.1.4.2.1. "A-towns" in Denmark. Sampling locations for drinking water (Table A.1.1.4 1.), bread (Table A.1.2.3.), vegetables and fruit (Tables A.1.2.5.1.) and A.1.2.5.2.), fresh dairy milk (Table A.1.3.2.), meat (Table A.1.3.4.1.) and total diet (Table A.1.4.2.). The "A-towns" were used in 1961-1972. The eight zones are indicated by Roman numerals.

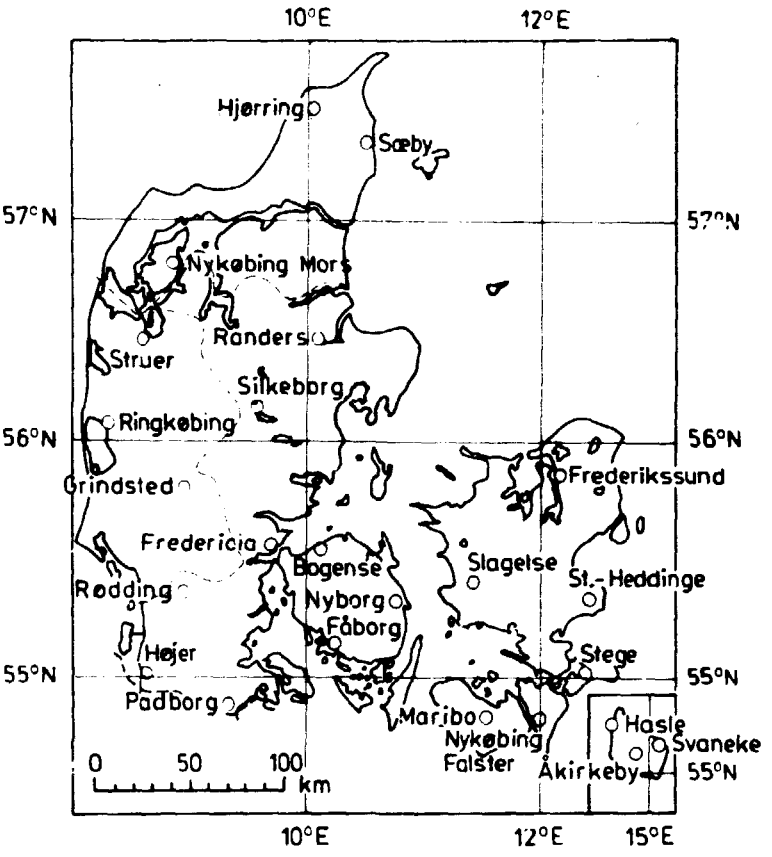

Fig. A.1.4.2.2. "B-towns" in Denmark. Sampling locations for drinking water (Table A.1.1.4.1.), bread (table A.1.2.3.), vegetables and fruit (Tables A.1.2.5.1.) and A.1.2.5.2.), fresh dairy milk (Table A.1.3.2.), meat (Table A.1.3.4.1.) and total diet (Table A.1.4.2.). The "B-towns" were used in 1963-1972.

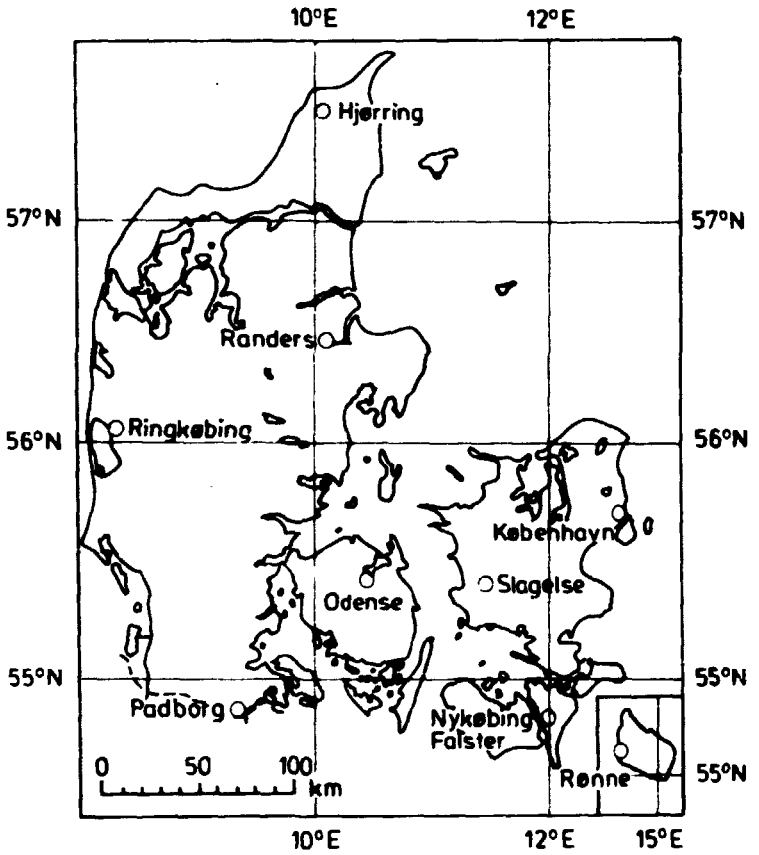

Fig. A.1.4.2.3. Sampling locations (since 1973) for bread (Table A.1.2.3.), fresh dairy milk (Table A.1.3.2.), and total diet (Table A.1.4.2.).

Table A.1.4.3. Sampling of human bone

	Danish human bone (RRD 61)	Faroese human bone (RRF 64)
Method	Samples of columna, preferentially from victimes of accidents. From the Institutes of Forensic Medicine in Copenhagen and Århus	Various bones from Dronning Alexandrine's Hospital in Thorshavn
Size	Adults : ~ 10-20 g Infants: a few grams	Usually as the Danish
Locality	The Institute in Århus receives samples from Jutland, and the Institute in Copenhagen from the Islands, mostly Zealand	Samples from the various parts of the Faroes were collected in Thorshavn
Period	Since 1961	Since 1964 (irregularly)
Frequency	Throughout the year (100-150 samples per year)	Running (2-3 samples per year)
Nuclides	^{90}Sr, Ca (Sr)	^{90}Sr, Ca
Pooling before analysis	If the samples contain more than 1 g ash they are not pooled. Bone from infants and especially from newborn children is occasionally pooled	Comparable samples containing less than 1 g of ash are pooled.

Table A.1.4.4. Sampling of deciduous teeth

	Danish teeth (XI)	Faroese teeth (XI)	Greenlandic teeth (XI)
Method	Teeth are collected by teachers or school dentists from pupils in smaller local schools. The teeth are placed in polyethylene bottles marked with the birth year of the donor. At Risø, the teeth are sorted according to type: incisors, cuspids, first and second molars.		
Size	2 - 10 g of tooth ash (depending upon the activity level) included in one analysis (1 crown yields 0.19 g ash on the average)		
Locality	Eighteen Danish schools (Fig. A. 1.4.4.)	Schools in Thorshavn, Tvøra and Klaksvik (Fig. A.1.1.3.2.)	Schools in Angmagssalik, Julianehåb and Godthåb (Fig. A.1.1.3.3.)
Period	1962-1975	Since 1962	Since 1962
Frequency	Throughout the year	Throughout the year	Throughout the year
Nuclides	^{90}Sr, Ca	^{90}Sr, Ca	^{90}Sr, Ca
Pooling before analysis	Teeth from the same area (Jutland, The Islands, Bornholm, The Faroes and Greenland), birth year, type and sampling year are pooled before analysis		
Remarks	Only crowns are included; fillings and roots are removed before analysis. Some samples of roots have been analyzed.		

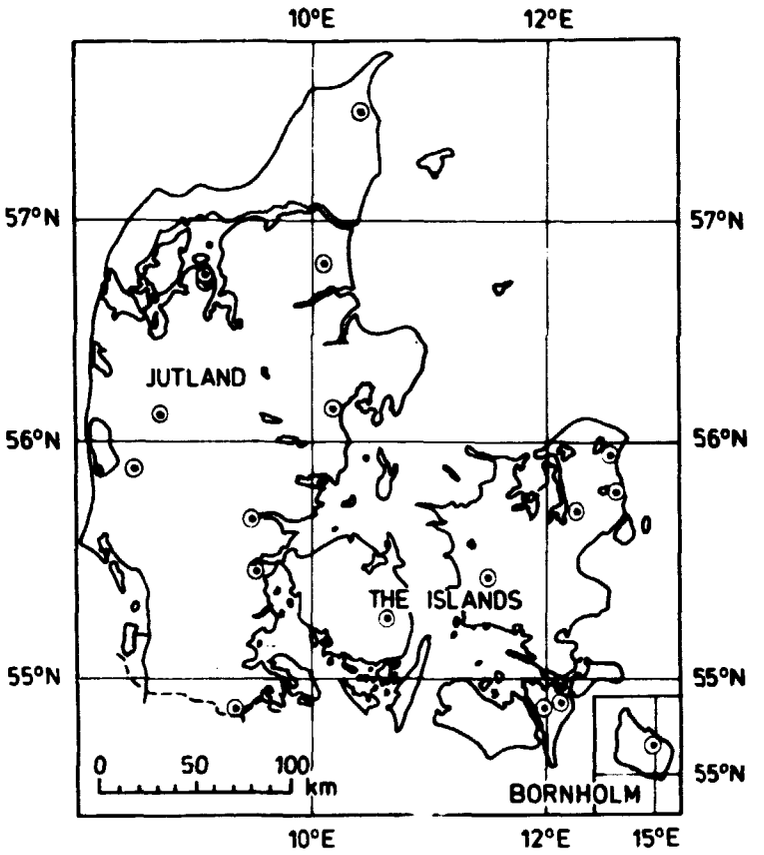

Fig. A.1.4.4. Sampling locations for deciduous teeth from pupils (Table A.1.4.4.).

Table A.1.4.5. Wholebody measurements

	Risø control group (RRD 63, RRD 70)	Faroese (RRF 63, RRF 69, RRF 74)	Greenlanders (RRG 74)	Donors of mothers' milk (RRD 64)
Method	Approx. 20 employees of the Health Physics Department at Risø were measured in a whole-body counter equipped with a 8x4 inch NaI crystal. The individual counting time was 1 hour	A few individuals were measured at Risø	Three individuals were measured at Risø	Three individuals were measured at Risø
Size	Approx. 20 persons (~ 10 males and ~ 10 females)	4 males and 4 females in total	1 male and 2 females in total	3 females
Locality	Roskilde, Zealand	Faroes	West Greenland	Zealand
Period	Since 1963	1963, 1969, 1974	1974	1964 (Dec. 1963-Aug. 1964)
frequency	3 times a year (March, August and December)	Once	Once	Twice
Nuclides	^{137}Cs, K	^{137}Cs, K	^{137}Cs, K	^{137}Cs, K
Pooling before analysis	Individual measurements	Individual measurements	Individual measurements	Individual measurements
Remarks	Until 1969, a 3x3 inch crystal was used			(cf. table A.1.4.6.)

Table A.1.4.6. Sampling of human milk

	Human milk from Zealand (RRD 62, XIII)
Method	Samples from individual donors who collected surplus milk not consumed by their babies. The milk was sent to Risø in polyethylene bottles
Size	3 donors supplied milk for 3/4 year (approx. 0.5 l d^{-1} per donor), 3 for 1/2 year, and 10 for a few months
Locality	Most donors were from North Zealand
Period	1962-1969 (RRD 69)
Frequency	The year round, but irregularly
Nuclides	^{90}Sr, ^{137}Cs, Ca, K
Pooling before analysis	Analysis of weekly or monthly samples for ^{137}Cs, usually monthly samples for ^{90}Sr
Remarks	Simultaneous sampling of donors' diet in 1962, 1964 and 1965 (Table A.1.4.2.) Wholebody counting of 3 milk donors in 1964.

A.2. Analytical procedures and measurements of radioactivity

Radioactive contamination is assessed by measurements of the ionizing radiation emitted as a result of the decay of the radionuclides. For γ-emitters, modern detector systems often permit a qualitative as well as a quantitative assessment, even of mixtures of several radionuclides, without any chemical operations. With respect to α- and β-emitters, radiochemical treatment before radiation measurement is usually warranted.

The aim of a radiochemical analysis is to separate a given radioelement from other interferring elements so that measurement of the radionuclide to be determined becomes possible. The general procedure consists of a removal or reduction of the sample matrix followed by various separations such as precipitation, extraction or ion exchange, or combinations of these operations. At the beginning of a radiochemical analysis, carrier and "spike" are added to the sample. The carrier is either the inert form of the element being analyzed, or a

non-isotopic chemical congener. The purpose of the carrier is to ensure that the extremely small chemical amounts of the radionuclide being determined are carried through the chemical operations. Furthermore, the carrier may be used for the determination of the recovery from the radiochemical analysis. However, yield is often more conveniently determined by means of a "spike", which is a radioactive tracer isotopic to the radionuclide in question. It is a prerequisite that the radiation of the spike does not interfer with the measurement of the sample itself. This may be achieved by using, e.g., a pure γ-emitter as spike for a pure β-emitter, or by applying γ- or α-emitters with energies different from those of the nuclide to be determined.

A.2.1. Radiostrontium analysis

The analytical procedures applied in this study for radiostrontium determinations were based on the methods developed at Harwell (Br59, Os59) and at the USAEC Health and Safety Laboratory (recently renamed Environmental Measurements Laboratory, U.S. Department of Energy) (Har172). The various procedures have been adapted as described in the Risø Reports (RRD59-76). The pretreatment of the various types of sample differs depending upon the matrix. Bone and rainwater samples only need minor treatment before fuming nitric acid separations (Fig.A.2.1.) that separate strontium from calcium and from most other elements, except barium and radium, because the nitrates of Sr (and Ba, Ra) are hardly soluble in 75% fuming nitric acid. Samples such as sea water and especially soil need laborious pretreatment before fuming nitric acid separation can be carried out. Two radioisotopes may occur in a radiostrontium analysis: 28-year*) ^{90}Sr (β = 0.54 MeV) and 50-day ^{89}Sr (β = 1.46 MeV); the nuclides are both pure β-emitters, which complicates simultaneous assessment. However, ^{90}Sr is determined indirectly measuring the 64-hour ^{90}Y (β = 2.3 MeV) daughter, and afterwards the count from the ^{90}Sr and ^{90}Y is subtracted from the total radiostrontium count, thus giving ^{89}Sr. The yield was assessed by the 65-day ^{85}Sr spike (γ = 0.514 MeV).

*) The estimates of the halflife of ^{90}Sr have varied throughout the years. The most recent estimate applied by Environmental Measurement Laboratory, New York is 28.82 y (Oct. 1980).

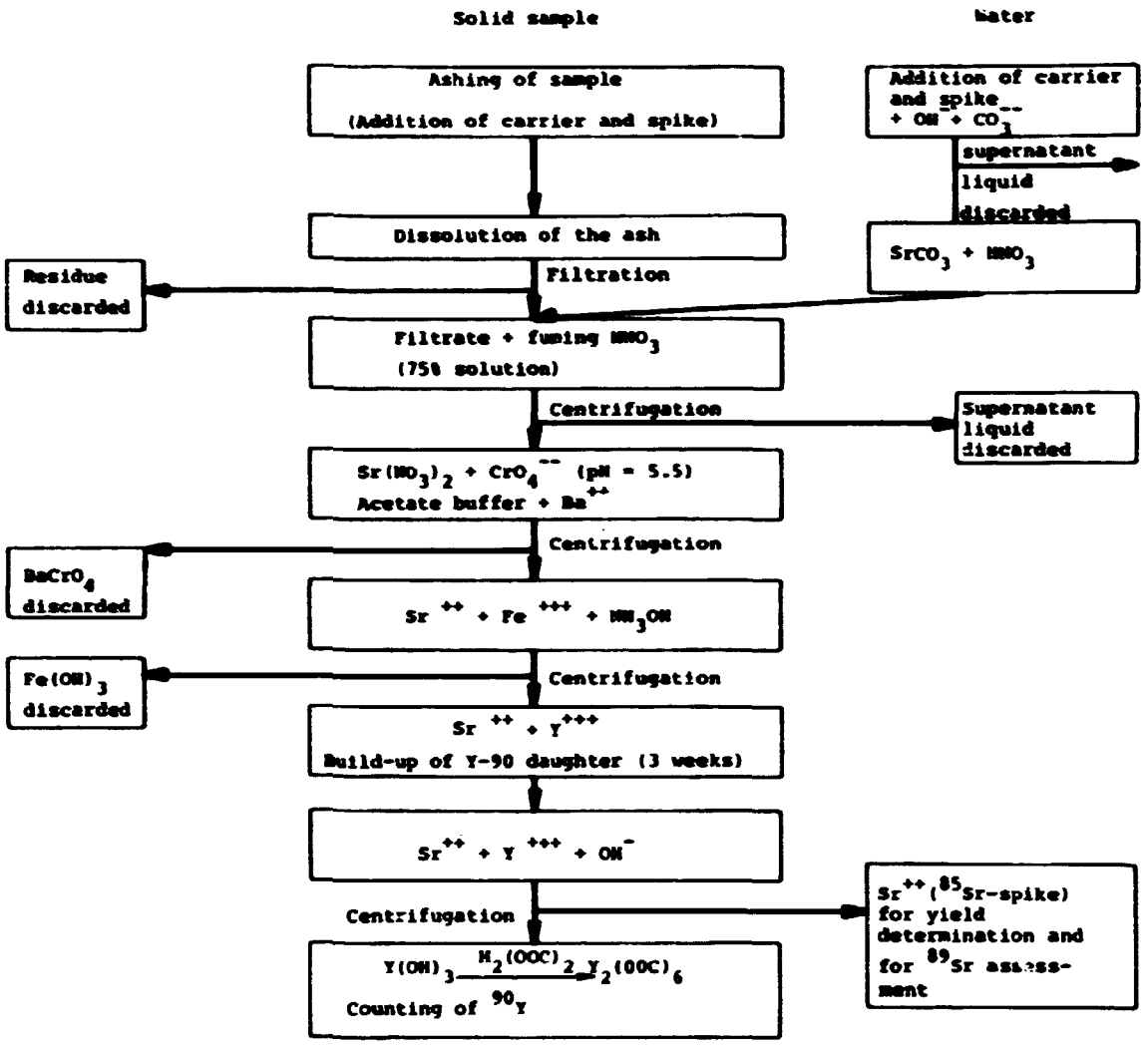

Fig. A.2.1. Sr-Analysis (fuming nitric acid method)

A.2.2. Plutonium analysis

The plutonium isotopes normally assessed in environmental samples have been $2.44 \cdot 10^4$ year ^{239}Pu ($\alpha \sim 5.14$ MeV), 6537 year ^{240}Pu ($\alpha \sim 5.16$ MeV) and 86.4 year ^{238}Pu ($\alpha \sim 5.49$ MeV). The first two isotopes are usually assessed as one nuclide: $^{239,240}Pu$, because the α-energies are too similar for individual determinations.

Two methods were applied for the determination of plutonium in environmental samples analysed at Risø. The first method used

(IV, Aa75) was a combination of a solvent extraction method developed by the Risø Chemistry Department and an ion exchange procedure used at the Oceanographic Institution at Woods Hole. This method was later (IX, V) replaced by a pure ion exchange procedure concluded by an electrodeposition of plutonium on a stainless steel planchet (Ta71) (Fig.A.2.2.). Plutonium was retained as an anion complex on an anion exchange resin, while interferring nuclides were eluted. Finally, plutonium was eluted from the resin by reduction of Pu^{+4} to Pu^{+3}, which disintegrated the complex. The yield was determined by the 2.85-year ^{236}Pu spike ($\alpha \sim 5.76$ MeV).

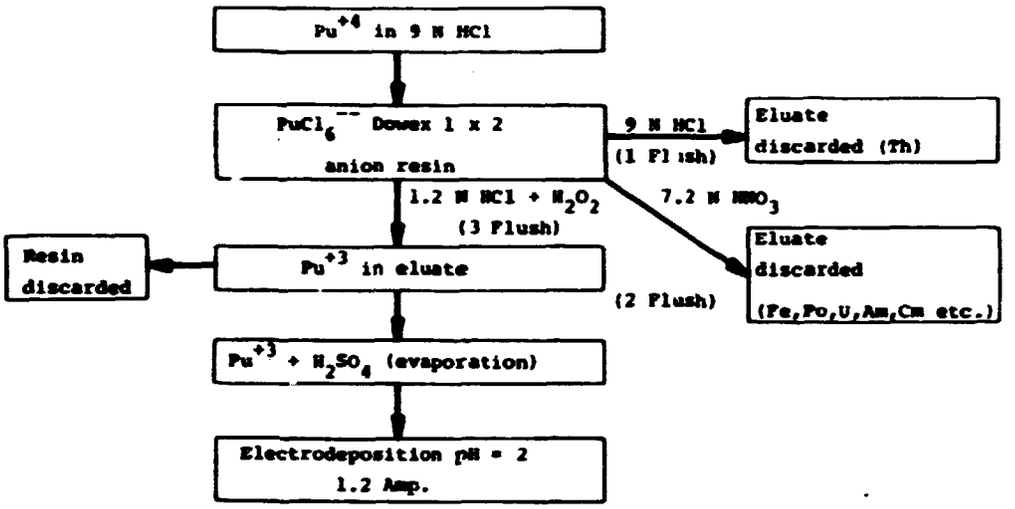

Fig. A.2.2. Pu-Analysis (after TALVITIE(Ta 71))

A.2.3. Assessment of radiocesium

The 2.55-min ^{137}Ba daughter of 30-year ^{137}Cs emits γ-radiation (0.662 MeV). As the mother and daughter in practice are in equilibrium, the γ-radiation is usually referred to as ^{137}Cs. If there is a sufficiently high ^{137}Cs concentration in a sample, the activity may be determined without any preconcentration. During the years showing high fallout rates, most samples were thus measured for ^{137}Cs as fresh or dried samples. When the levels decreased at the end of the sixties, it became necess-

ary to reduce the matrix for most samples. This was accomplished by drying or ashing; as ^{137}Cs may evaporate at higher temperatures (Harl72), the ashing temperature was kept at 400°C. For sea water, the radiocaesium was collected on an AMP (ammonium molybdate phosphate) precipitate (Du70), which acted as a cation exchange medium. The AMP was stirred in the sea water for one hour, and then the water was drawn off after the AMP had settled. This method recovered approx. 95% of the radiocaesium present in sea water. The recovery of ^{137}Cs was determined with the 2.06-year ^{134}Cs spike (γ = 0.796 MeV).

A.2.4. Radioactivity measurements

The concentrations of radionuclides in environmental samples are in general low. Hence the detector systems used for the measurements of various types of radiation should have both high efficiency and low background. In other words, the systems should be fitted for low level counting. In an earlier study (Li63), LIPPERT described the various systems applied at Risø for the measurement of α-, β- and γ-emitters in environmental samples. Since LIPPERT wrote his treatise in 1963, semiconductor detectors have been developed (Li68) and these have increasingly been used for α- and γ-spectroscopy. Table A.2.1 summarizes the various methods used in this study for the measurement of ionizing radiation in environmental samples.

Alpha and gamma emitters are assessed through spectral data analysis usually carried out by means of a computer using a least squares fit (Harl72). The background and contributions from other nuclides are subtracted from the counts in the photopeak of the radionuclide being determined. In the case of spectra obtained by solid state detectors, the quality of the spectra often permits a more simplified assessment of a radionuclide. The background counts are determined on both sides of a photopeak and the net count in the peak is calculated by subtraction of the background thus estimated (Li75). If, however, several nuclides are present, or if the activity levels are low compared to the background count, the least squares method is preferable for spectral analysis. This method is in general always used for NaI(Tl) γ-spectra, because of the poor spectral resolution of these detectors.

Table A.2.1. Counting systems in use at Risø for environmental samples

System No.	Radiation	Nuclide	Efficiency cpm per pCi	Background cpm	Sample geometry and sample material
1[a]	α	^{239}Pu	0.62	0.026	20 mm Ø electroplated stainless steel planchet
2[a]	β	^{90}Y	0.87	1.5	20 mm Ø filter with ~ 30 mg yttrium oxalate. Mounted on nylon disk (Harl 72)
3[a]	β	^{90}Y	0.94	0.28	20 mm Ø filter with ~ 30 mg yttrium oxalate. Mounted on thallium disk
4[a]	β	^{90}Y	0.74	0.29	20 mm Ø filter with ~ 30 mg yttrium oxalate. Mounted on thallium disk
5[c]	γ	^{137}Cs	0.0038	0.21	50-100 g ash in a 80 mm Ø x 45 mm cylindrical can
			0.0031	0.27	~ 1 kg in a 1.7 l can ("hat geometry" [b]) (dried milk)
			0.0028	1.11	~ 2.5 kg in a 1.7 l can ("hat geometry" [b]) (soil)
			0.0064	0.29	50 g AMP in 87 mm Ø x 8 mm cylindrical perspex container
6[c]	γ	^{137}Cs	0.0154	0.20	12 x 15 x 1 cm^3 folded glass fiber filter (35 g)
7[d]	γ	^{137}Cs	0.0067	57	70 kg "standard" man

a) The Geiger flow counters had a 1.8 mg cm^{-2} mylar foil window.

b) The cylindrical sample container fits over the detector house.

c) Before the Ge(Li) detectors became available, the γ-measurements were carried out on 3" x 4" NaI(Tl) crystals.

d) The person being measured sits in a so-called "tilted-chair" geometry (Ia 64). The chair is placed underneath the detector in a cylindrical (2m Ø x 2 m) room ventilated by filtered air. The room is the gun-tower of the Danish frigate "Peder Skram", which was scuttled by the Danish Navy on 29 August 1943; the iron in the tower is thus free of ^{60}Co.

Number of detectors	Detector	Auxiliary equipment
10	450 mm^2 silicon diode Ortec	Nuclear Data multichannel analyzer. 64 channels per detector. Detector in vacuum chamber.
5	40 mm ϕ Geiger flow counter; made at Risø [a]	Each detector shielded by 75 cm iron and 2 cm lead; automatic sample changers for 30 samples
5	25 mm ϕ Geiger flow counter made at Risø	Each detector shielded by 10 cm lead; automatic sample changers for 4 samples
3	25 mm Geiger flow counter made at Risø	One detector shielded by 15 cm iron. Two detectors each shielded by 10 cm lead. Manually operated
4	~ 40 cm^3 closed-end, coaxial type, lithium-drifted Ge semiconductor detectors; made at Risø	Each detector is connected to a 1024-channel analyzer (3 Hewlett Packard and 1 Nuclear Data). The detectors are shielded by 10 cm lead [c]. (The background count was 0.11 cpm in the ^{137}Cs peak if there was no sample).
1	~ 100 cm^3 coaxial Ge(Li) detector Canberra	1024-channel Canberra analyzer detector placed in wholebody shielding (18 cm iron) surrounded by 5 cm lead
1	8" x 4" NaI(Tl) scintillation crystal Harshaw	1024 TMC channel analyzer Wholebody shielding; 18 cm iron + 1 cm lead + 1 mm Cd [d]

A computer program is also used for the calculation of ^{90}Sr based on the counting of the ^{90}Y daughter when equilibrium has been attained after a buildup period of approx. 3 weeks. The yttrium sample is usually counted in 6 periods of 180 minutes for 3-4 days in an automatic sample changer. The program subtracts the background count and calculates the count-weighted mean and the relative standard deviation of the six countings which have been decay-corrected back to the time of separation of the 64-hour ^{90}Y daughter from the ^{90}Sr mother solution. From information on counter efficiency and on the recovery of strontium and yttrium in the radiochemical analysis, the ^{90}Sr concentration of the sample is calculated.

The sensitivity of the various detector systems depends on the counter efficiency, including sample geometry and background. The sensitivity is described by the so-called lower limit of detection (LLD) (Pa71). The lower limit of detection may be expressed by a simplified formula (Har172) as:

$$LLD = 2\ Ks\ \sqrt{2} \qquad \text{(Eq.A.2.1.)}$$

where s is the estimated standard error of the background count, and K is the value for the upper percentile of the standardized normal variate, corresponding to a preselected risk of falsely concluding that activity is present (α).

Equation A.2.1 furthermore implies that one also accepts the chance of detecting activity when it is present (100 - α) % of the time, but of failing to detect it α% of the time. If α = 5%, K becomes 1.645 and LLD = 4.66 · s (Har172). The lower level of detection may also be expressed by the minimum activity detectable in a given sample by a given procedure:

$$LLD = \frac{2\ K\ \sqrt{2} \cdot s}{E \cdot Y} = \frac{4.66 \cdot s}{E \cdot Y} \quad \text{(if } \alpha = 5\%\text{)} \qquad \text{(Eq.A.2.2.)}$$

where E is the counter efficiency in cpm per pCi, and Y is the chemical recovery of the nuclide in question by means of the procedure applied.

Table A.2.2 summarizes LLD values for the most common types of environmental samples analyzed at Risø.

Table A.2.1: [illegible]

[illegible]	^{90}Sr [a]	^{137}Cs [b]	$^{239,240}Pu$ [c]
[illegible]	[illegible]	[illegible]	[illegible]
Air filter	[illegible]	[illegible]	
Rain water	[illegible]		
Ground water	[illegible]		
Sea water	[illegible]	[illegible]	[illegible]
Soil	[illegible]	[illegible]	[illegible]
Forest grass	[illegible]	[illegible]	[illegible]
Milk	[illegible]	[illegible]	
Total diet	[illegible]	[illegible]	
Bone	[illegible]		

[a] The mean recovery of the ^{90}Sr analysis was 80%. The detector was system No. 3 in table A.3.1. The food samples (grain, milk and total diet) were analysed on 10 g ash. The average count of a blank was 0.05 cpm; this contribution was included in the estimate of LLD for ^{90}Sr.

[b] All samples were counted on system No. 5 in table A.3.1. except the air filter which was measured on No. 6.

[c] The average recovery in Pu analysis was 70%; however in Pu analysis on sea water the recovery was 90% only.

[d] 100 l samples of distilled water and ion-exchanged water showed 0.0 ± 0.7 dpm (1σ) per sample (7 samples analysed 1960-1970).

A.3. Quality of data assessment

An inherent difficulty in an environmental study extending over nearly two decades is the inevitable alteration in the procedures applied. The staff who collect and analyze the samples change throughout the years, and so to some extent do the procedures for the physical and chemical treatment of the samples. The methods of measurement and calculation have also been subject to modifications. These alterations may have unintentionally influenced the time variation of the material in particular. Hence in all analytical work concerning environmental long-term studies it is of special importance to check the precision as well as the accuracy of the data produced.

A.3.1. The precision of the data

The precision, or the reproducibility of the determinations, is defined as the extent of agreement between individual values. Precision is thus a measure of the quality of the measurements. However, precision gives no information on a possible bias. It is thus possible to produce results of high precision but of low accuracy if, e.g., the standards used for the calibration of the counting systems are in error.

The counting error is used to measure the quality of the physical measurements, and it is estimated as the Poisson standard deviation. If the count rate of a sample is T counts in n minutes, and the background is B counts in m minutes, then the net count rate becomes:

$$N = \frac{T}{n} - \frac{B}{m}$$

and its standard deviation (Fr57):

$$s_n = \sqrt{\frac{T}{n^2} + \frac{B}{m^2}} .$$

In the case of single determinations, it has been the practice to use one s_n as an estimate of the precision. However, this underestimates the overall error of a determination because it includes neither errors from chemical operations nor from sampling. A better estimate is one standard error (SE) of the mean obtained from double or multiple determinations. Of the approx. 15 000 ^{90}Sr determinations in our material, 15% or approx. 2 300 analyses were carried out as duplicates. The double determinations were performed in different laboratory rooms by different analysts. To test whether systematic changes in the analytical performances had occurred throughout the years, a set of so-called blind duplicates (harl72) was analyzed in 1977. Unknown to the analysts, the samples were old samples that had been analyzed for ^{90}Sr earlier. From table A.3.1 it appears that the precision for the blind duplicates corresponded to that of the usual duplicates. On the average, the precision was approx. 9%; this came close to the mean %

Table A.3.1. The precision of ^{90}Sr determinations, estimated from usual and blind duplicates. The precisions were given as mean % deviations, equal to the relative standard error (as percentage) of the mean of the duplicates [a].

Sample	The precision estimated from usual duplicates (± 1SE)	Numbers of duplicates included [b]	The precision estimated from blind duplicates collected 1963-1975; range in brackets	Numbers of blind duplicates
Soil	(6.7 ± 0.8) %	100	2.9% (0.2-5.7%)	4
Grain	(6.9 ± 0.6) %	100	6.1% (1.6-10.8%)	4
Grass	(10.0 ± 1.9)%	46	8.9% (6.1-15.0%)	4
Potatoes	(9.5 ± 0.9) %	91	-	-
Sea plants	-	-	15.4% (8.4-31.6%)	4
Dried milk	(6.6 ± 0.6) %	100	4.0% (1.1-7.7%)	4
Animal bone	-	-	11.5% (0.5-20.0%)	8
Meat	-	-	13.8% (7.9-20.2%)	4
Total diet	(5.7 ± 0.4) %	90	-	-

a) Let the two determinations in a double analysis be A and B, hence the mean is $\frac{A + B}{2}$ and the mean % deviation from the mean is $\frac{A - B}{A + B} \times 100$, which equals the relative SE (as percentage) = $[100 \cdot ((A-(A+B)\cdot 2^{-1})^2 + (B-(A+B)\cdot 2^{-1})^2)^{0.5} \cdot 2^{-0.5}] \cdot [(A+B)\cdot 2^{-1}]^{-1}$.

b) Randomly selected among all duplicates analyzed since 1959, if the number was less than 100, all determinations were included.

deviation of ^{90}Sr quality control samples analyzed by the U.S. Health and Safety Laboratory (HASL) (Harl72). For blind duplicates of bone and vegetation analyzed in 1962-1975, HASL found a mean deviation of 10%.

The sampling error was estimated for air filters, precipitation, various diet components and total diet as shown in table A.3.2. For air and precipitation, the estimates also included a possible difference in the performance of the two sets of sampling systems applied. For vegetables a possible time variation during the sampling month was included; for wheat, there was a contribution from differences between winter and spring varieties. The estimated precision of sampling and analysis of ^{90}Sr in the total diet did not differ whether the samples were collected in the two groups of towns (A & B) within

Table A.1.2. Precision of sampling estimated from double samplings. The precisions are given as mean % deviations

Sample and location	Number of duplicates	Nuclide	Precision of sampling and analysis (±1SE) (T)	Estimated precision of analysis (A)	Estimated precision of sampling $S = \sqrt{T^2 - A^2}$	Sampling methods for the duplicates
Air at Risø	131	^{90}Sr	(14.5 ± 0.9)%	~ 9%	~ 11%	The small and the large air sampler 1964-1975 (cf. table A.1.1.2.)
Precipitation at Risø	105	^{90}Sr	(17.9 ± 1.2)%	~ 9%	~ 15%	The ion exchange columns and the rain bottles at Risø 1962-1975 (cf. table A.1.1 3.1.).
Wheat from state exp. farms (fig. A.1.1.3.1.)	107	^{90}Sr	(10.4 ± 0.7)%	6.9%	~ 8%	Spring and winter varieties, 1962-1971 (cf. table A.1.2.2.)
	56	^{137}Cs	(10.3 ± 1.2)%			
Cabbage from 8 zones (fig. A.1.4.2.1. & A.1.4.2.2.)	24	^{90}Sr	(25 ± 3)%	10%	~ 23%	A and B towns in the 8 zones 1969, 1970 and 1972. (cf. table A.1.2.5.1.)
Leaves of brussels sprouts from test growing stations (fig. A.1.2.5.)	16	^{90}Sr	(12 ± 2)%	~ 10%	~ 7%	The samples were collected in the first and second half of the months in 1962-1963 (cf. table A.1. 2.5.2.)
Fresh dairy milk 8 zones (figs. A.1.4.2. 1. and A.1.4.2. 2.)	16	^{90}Sr	(7.3 ± 1.6)%	6.6%	~ 3%	A and B towns in the 8 zones in Sept. and Dec. 1963 (cf. table A.1.3.2.)
	16	^{137}Cs	(6.1 ± 1.4)%			
Beef and pork 8 zones (figs. A.1.4.2.1. and A.1.4.2.2.)	16	^{137}Cs	(13 ± 3)%			A and B towns in the 8 zones in Dec. 1963 (cf. table A.1.3.4.1.
Total diet 8 zones (figs. A.1.4.2.1. and A.1.4.2.2.)	151	^{90}Sr	(8.3 ± 0.6)%	5.7%	~ 6%	A and B towns 1963-1972 (cf. table A.1. 4.2.)
	151	^{137}Cs	(9.7 ± 0.7)%			
Total diet 8 zones (fig. A.1.4.2.)	30	^{90}Sr	(8.6 ± 0.9)%	5.7%	~ 6%	A towns only: two groups of shops 1961-1962

The overall precision may also be estimated from the residual error in the anova (cf. B.) However, the anova error is the relative standard deviation. Hence, when comparing the above estimates of precision from sampling and analysis with those obtained from the anova, the former should be multiplied by $\sqrt{2}$.

the zones (figs.A.1.4.2.1 and A.1.4.2.2), or whether they were collected in two groups of shops in the A towns only. The overall mean precision of sampling and analysis was from tables A.3.1 and A.3.2 estimated at 10-15% for ^{90}Sr corresponding to a relative standard deviation of about 15-20%; however, it was evident that the error varied with the type of sample.

In the case of ^{137}Cs, the quality of the determinations was estimated by recounting samples previously counted as fresh samples on NaI(Tl) crystals. The recounting was performed on ash on Ge(Li) detectors. The mean % deviation between old and new determinations was 12% for 25 samples of grain, milk and grass counted the first time in 1963-1968 and the second time in 1971 and 1972. The study showed no significant difference between the new and the old determinations, which suggested that the ashing applied to the samples did not influence the ^{137}Cs content of the samples systematically (cf.A.2.3). The determinations of the precision of sampling and analysis of ^{137}Cs in grain, milk and total diet in table A.3.2 suggested that the analytical precision of ^{137}Cs determinations was generally better than the 12% obtained from the recounting; the analytical precision for ^{137}Cs was estimated from table A.3.2 to equal that of ^{90}Sr.

A.3.2. The accuracy of the data

The accuracy or overall error of a measurement is defined as the extent of agreement between the observed and the true value. Strictly speaking, the accuracy can only be determined by measurements of standard samples of the same material as the sample being determined. This is in general difficult because environmental standard samples usually have a fallout background, unless they were obtained prior to nuclear weapons testing. To reduce the influence of a possible background in the standard samples, these are spiked with relatively large amounts of radionuclides; this, however, depreciates their value as standards for low level analysis. Accuracy can, however, also be assessed by comparing the results of a large number of analyses obtained by several, experienced laboratories on so-called intercomparison samples. These samples are "naturally" contaminated (by fallout or releases from nuclear installations) and the true concentrations of radionuclides are thus unknown.

Throughout the years the International Atomic Energy Agency (IAEA) (Me69) in Vienna and Monaco and the Health and Safety Laboratory

Table A.1.1. Intercomparison samples analysed at Risø in 1960-1977. The figures are the mean ratios between the Risø determination and the recommended value of the sample; the number of samples is shown in brackets; a indicates spiked samples.

		1960-61	1962-63	1964-65	1966-67	1968-69	1970-71	1972-73	1974-75	1976-77
Air filters	^{90}Sr							1.06(10)a		
	^{137}Cs							0.97(10)a		
	^{239}Pu							1.00(4)a		
Sea water and other water samples	^{90}Sr		1.02(1)		1.19(3)		0.86(1)	1.00(1)a	1.38(1)a	1.13(2)
					1.26(1)a		1.31(1)	0.98(1)a	1.19(1)a	1.26(1)
					1.06(1)a					
	^{137}Cs						-	0.67(1)a	1.12(1)a	1.26(2)
							1.11(1)	0.89(1)a	0.90(1)a	1.01(1)
	^{239}Pu						0.93(1)		1.16(1)a	
							0.99(1)		1.11(1)a	
Soil	^{90}Sr	1.07(2)		0.99(5)						
		1.08(2)								
Sediments	^{90}Sr							1.02(1)		
	^{137}Cs							1.03(1)		
	^{239}Pu							1.05(1)		
Cereals	^{90}Sr		1.17(3)	0.98(6)						
	^{137}Cs				1.02(3)					
Various vegetation	^{90}Sr			1.03(6)				0.93(1)		
				1.27(1)						
	^{137}Cs							0.91(1)		
Milk	^{90}Sr		1.21(4)	1.24(2)		1.13(6)			1.01(7)	
	^{137}Cs					0.96(2)			1.09(2)	
Various animal samples	^{90}Sr		0.82(4)	0.72(2)					1.58(1)	
				1.06(8)					0.30(1)	
	^{137}Cs			1.01(2)					1.11(1)	
									1.12(1)	
	^{239}Pu								1.36(1)	
Mixed diet	^{90}Sr			0.99(6)						
	^{137}Cs			0.90(1)						
Bone	^{90}Sr		0.93(4)	1.03(12)					1.04(6)	0.97(8)

The mean ratio of 123 ^{90}Sr intercomparison ratios was 1.04±0.13 (15%)
" " " " 32 ^{137}Cs " " " 1.01±0.11 "
" " " " 10 $^{239,240}Pu$ " " " 1.02±0.19 "

(HASL) (Ha58-78) in New York have organized intercomparison programmes. Since 1960 Risø has participated in these programmes and table A.3.3 summarizes the results of ^{90}Sr, ^{137}Cs and $^{239,240}Pu$ intercomparisons. It is evident that Risø's results have not shown any time trend in accuracy, nor have the results taken as a whole deviated significantly from the means of those considered by the IAEA and HASL as the "best values" in these intercomparisons. It is not a serious matter if the true value for the activity content in environmental samples shows minor deviations from that obtained through the intercomparisons; it is more important that the results from the various laboratories engaged in these environmental studies are unbiased in relation to one another. Table A.3.3 suggests that the coefficient of variation for Risø's results compared to the "best value" was 13% for ^{90}Sr, 11% for ^{137}Cs and 19% for $^{239,240}Pu$. HASL (Har172) found a relative standard deviation of replicate analyses of reference samples (bone, milk, vegetation) of 11% for ^{90}Sr.

APPENDIX B

ANALYSIS OF VARIANCE, AND THE VARIABILITY

This appendix contains a summary of the principles for analyses of variance (anova) and the definitions for the statistics used throughout the text; for details, reference should be made to text-books in statistics (Ha57,Da56,So69). Furthermore, examples are given of anovas of the various sample categories and calculations of the so-called variability.

B.1. Introduction

Both in radioecology and in other environmental and biological sciences, many causal factors, uncontrollable in their variation and often unidentifiable, affect most phenomena. One needs statistics to measure such variable phenomena and to ascertain the validity of very small but perhaps important differences.

During the first period of operational environmental monitoring at Risø, Professor A. HALD was approached for advice concerning the treatment of the data relating to the measurements performed. Professor HALD pointed out the analysis of variance as a very suitable statistical tool for the evaluation of the significance of the observed variations of the data with time, location and species. Analyses of variance were consequently introduced at an early stage (RRD58-59) of the present study as the statistical method for data treatment.

B.2. Some statistical definitions

A sample of observations is a collection of individual observations selected by a specified procedure.

The actual property measured by the individual observations is the variable and a single observation of a given variable is called a variate.

A population is the totality of individual observations about which inferences are to be made, existing anywhere in the world, or at least within a definitely specified sampling area limited in space and time.

Normally, interest is centered on the populations from which the samples have been taken. It may, e.g., be the population mean or the population variance. These so-called population statistics or parameters are, however, usually unknown and instead the so-called sample statistics are applied. Thus the sample mean $\bar{x}$ estimates μ, the parametric mean of the population, and a sample variance s^2 estimates the parametric variance σ^2. Such estimators should be unbiased, i.e. an infinite number of samples taken from the population should give sample statistics that, when averaged, will give the parametric value.

The variance or the mean square is the sum of squared deviations divided by the number of terms:

$$\sigma^2 = \frac{\sum_{i=1}^{i=n} (\mu - x_i)^2}{n} \qquad \text{(Eq.B.1)}$$

where x_i is an individual observation.

The corresponding sample variance is defined as:

$$s^2 = \frac{\sum_{i=1}^{i=n} (\bar{x} - x_i)^2}{n - 1} = \frac{SSD}{n - 1} \qquad \text{(Eq.B.2)}$$

where n - 1 is called degrees of freedom (f). If n had been applied instead of n - 1, s^2 would underestimate σ^2. To overcome this bias, the sum of squared deviations (SSD) is divided by n - 1.

The standard deviation is the square root of the variance:

$$SD = \sqrt{s^2} \qquad \text{(Sq.B.3)}$$

and the standard error of the mean $\bar{x}$ of n individual observations is:

$$SE = \sqrt{\frac{s^2}{n}} = \frac{SD}{\sqrt{n}}. \qquad \text{(Eq.B.4)}$$

Either SD or SE are used as the error term throughout the text. In each case it is stated which of the two statistics is applied. SD was preferred if interest was focussed on the variation of the individual values of the population on which the sample mean was based, while SE was chosen if the precision of the mean was considered to be more important.

The coefficient of variation is a statistic often used in the comparison of amounts of variation in populations having different means. It may be given as a percentage, but here it is quoted as a decimal:

$$CV = \frac{\sqrt{s^2}}{\bar{x}}. \qquad \text{(Eq.B.5)}$$

(The parametric value of CV is termed $\eta = \frac{\sqrt{\sigma^2}}{\mu}$).

As the coefficient of variations of radioactivity concentrations in environmental samples were relatively constant, the distribution of the data dealt with in radioecology was usually log normal (Eb64) (cf. also p 804-805 in ref. XII). Before statistical treatment, such as the anova, where additivity is assumed, the data were consequently transformed to their natural logarithms (ln). However, in accordance with other studies (Un58-77), the sample means were calculated as arithmetic means, unless anything else was stated. The logarithmic transformation implied that, for minor variances, η with approximation equalled the standard deviation σ of the transformed data (Ai69) as:

$$\eta = (e^{\sigma^2} - 1)^{\frac{1}{2}}. \qquad \text{(Eq.B.6)}$$

The estimator of the coefficient of variation CV was obtained by replacing σ^2 by s^2 in Eq.B.6.

The calculated values (Y_c) obtained from the prediction models (cf.C) were evaluated by a comparison with the corresponding observed values (Y_o), by means of the correlation coefficient:

$$r = \frac{\Sigma Y_c Y_o - \Sigma Y_c \Sigma Y_o/n}{\sqrt{(\Sigma Y_c^2 - (\Sigma Y_c)^2/n)(\Sigma Y_o^2 - (\Sigma Y_o)^2/n)}} \qquad \text{(Eq.B.7)}$$

where n is the number of data sets to be compared; the numerator in Eq.B.7 is the covariance of Y_c and Y_o, and the denominator is the square root of the product of the variances of Y_c and Y_o.

The correlation coefficient can range from +1 for perfect positive association to -1 for perfect negative association. The most common significance test is whether a sample correlation coefficient could have been derived from a population with a parametric correlation coefficient of zero. This null hypothesis was tested as a t-test with n-2 degrees of freedom:

$$t = r\sqrt{\frac{n-2}{1-r^2}}. \qquad \text{(Eq.B.8)}$$

B.3. Analysis of variance

The analysis of variance is a technique for statistical analysis. It tests whether two or more sample means could have been obtained from populations with the same parametric mean with respect to a given variable. Alternatively, it can be concluded that these means differ so much from each other that it must be assumed they were sampled from different populations.

In an anova the variates are grouped according to separate kinds of treatment. If only one source of variation is dealt with, the analysis is a one-way anova, in the case of two factors a two-way anova, etc.

As an example, a three-way anova table B.2.2.1 is considered. The variable is the natural logarithm of the ^{90}Sr concentration in pCi ^{90}Sr (g Ca)$^{-1}$ in the four species of grain collected at harvest in the period 1959 to 1974 at the eleven state experimental farms. In this case there are three factors, or treatments: Years (Y), Species (S), and Locations (L). These treatments are each responsible for a so-called main effect.

Because of the varying contamination levels of the atmosphere and the soil with time, the years will thus influence the annual grain levels. As a result of morphological and physiological characteristics, the species will determine the interspecific variation of the direct and indirect contamination of the grain, and finally the various locations will result in local variations of the variable because of differences in precipitation, soil characteristics and agricultural practice.

The structure of variation may be described as:

$$x_{hijk} = \mu + y_h + s_i + l_j + (ys)_{hi} + (yl)_{hj} + (sl)_{ij} + (ysl)_{hij} + \varepsilon_{hijk} \qquad \text{(Eq.B.9)}$$

where x_{hijk} is a single variate (the k determination from the year h, of the grain species i, at the location j); μ is the parametric grand mean of the population (the mean ^{90}Sr concentration of all grain grown at the eleven state experimental farms from 1959 to 1974; y_h, s_i, l_j are the treatment effects for the h'th, i'th and j'th group of treatments Y, S and L, respectively; $(ys)_{hi}$, $(yl)_{hj}$ and $(sl)_{ij}$ are the so-called first-order interaction effects in the subgroups represented by the indicated combinations of the h'th group of factor Y, the i'th group of factor S, etc.; $(ysl)_{hij}$ is the second-order interaction effect in the subgroup representing the h'th, i'th and j'th groups of factors: Y, S and L, respectively; and

ε_{hijk} is the error term of the k'th item in subgroup hij.

An interaction between two treatment factors may be exemplified by the interaction between years and locations (YxL). If this interaction happens to be significant, it means that the ^{90}Sr concentrations at the various locations did not follow the same time pattern. In certain years, location A may thus show higher levels than location B, while in others the opposite could be the case. The reasons for such interactions could be numerous, and they are often illustrative of the complex conditions in nature. Second-order interactions are less transparent than first-order. A second-order interaction means that the first-order interactions, e.g. between years and locations, may vary with species. An example of a second-order interaction is given in table 2.2.1. in the main text.

The principles of calculation of an anova are available in text-books and are not repeated here, but examples of the final tables of the anovas are contained in this appendix.

The significance test of a treatment - or an interaction effect - in an anova is carried out by a comparison of the variance estimate s^2 of the treatment or interaction in question with the error variance. A "null hypothesis" is made that all effects in the table of the anova have a zero magnitude, so that all the variance estimates (s^2) are independent estimates of the same quantity estimated by the error variance, i.e., an estimate of the magnitude of the experimental error. The so-called v^2 test evaluates whether the variance estimate (s^2) based on any of the possible sources of variation is so much greater than the variance estimate based on the experimental error, that the observed variance ratio (v^2) is unlikely to have arisen by chance.

In the case of significant interactions the anova may be split up; the above three-way anova may thus be split into four two-way anovas each representing one of the four grain species, or into eleven two-way anovas, one for each of the eleven farms, etc. The two-way anovas may further be divided up into one-way anovas. Such a "breakdown analysis" makes it possible

to test all sources of variation, but a little of the general view is lost.

The probability P in the anova table is the so-called probability fractile of the distribution in question (Ha57) - here the v^2 distribution (also called the F-distribution, but if so the probability is usually termed p; $p = \frac{100-P}{100}$). Throughout this work, the following terminology is used:

$P \geq 95\%$, $(p \leq 0.05)$: probably significant difference: *
$P \geq 99\%$, $(p \leq 0.01)$: significant difference: **
$P \geq 99{,}9\%$ $(p \leq 0.001)$: highly significant difference: ***

A significance level p is the probability of the result having arisen entirely by chance, the treatments having had nothing to do with it; e.g., $p \leq 0.01$ ($P \geq 99\%$) implies that the observed difference in one per cent (or less) of the cases may have been accidental.

In the significance tests of the anovas the "null hypothesis" was rejected, i.e. an effect was considered to be significant only if $P \geq 99\%$. An attempt is thus made to minimize the so-called type I error, which implies a rejection of a true null hypothesis, but this involves the risk of accepting a false null hypothesis, i.e. a type II error. However, in the present context it was considered more important to avoid type I errors, because one purpose of the study was to demonstrate essential differences in the radioactive contamination of the environment.

In an environmental study, as well as in many biological experimental studies, it is inevitable that some data will be missing, in such cases the anova becomes more complicated to perform. However, electronic data processing has made anovas feasible even when a relatively large number of data are lacking. The general principle has been described by DAVIES (Da56). Missing values are calculated by a least squares method in order to minimize the influence of the estimated figures on the residual error in the anova. VESTERGÅRD (Ve64) developed a computer program VAR-3 that in principle calculates the missing

figures as described by DAVIES, but VAR-3 does more because it can also produce a modification of the existing figures in such a way that the interactions are eliminated. The program, which has been adapted to the STATDATA program (Li75), thus produces a table showing the same tendencies regarding each main effect. Therefore the ratios between the levels found in two given years are the same for all locations and species, and the ratios between two species are the same for all years and locations, etc. These artificial numbers can be used for different purposes. If the interactions are insignificant, or not considered pertinent, the VAR-3 table may be used for depicting the main effects relative to the grand mean in column diagrams as shown throughout the text; the relative values in these column diagrams were calculated as arithmetic means. The modified data obtained by VAR-3 were used in a few prediction models (cf.tables C.3.6.2 and C.4.4.1), where some periods were less represented by certain samples than others. In these cases the VAR-3 data were preferred to the raw data because the VAR-3 recalculation minimized the shortcomings of the original data.

B.4. Variability

If the sums of deviations from all n sources in an anova (cf. e.g. table B.2.2.1.) are summed up and divided by the sum of the degrees of freedom (f_t), the total variance in the entire sample due to all causes is obtained:

$$s_t^2 = \frac{\sum_{i=1}^{i=n} SSD_{(i)}}{f_t} . \qquad \text{(Eq.B.10)}$$

s_t^2 is denoted the total variance around the grand mean; s_t^2 may be used for a calculation of the total coefficient of variation around the grand mean, hence in an anova, where the data are transformed to their natural logarithms:

$$CV_t = (e^{s_t^2} - 1)^{\frac{1}{2}}. \qquad \text{(Eq.B.11)}$$

EBERHARDT (Eb64) used the coefficient of variation as a measure of the variability of ^{90}Sr and ^{137}Cs in environmental samples. Instead of CV_t it may, however, be more rewarding to consider the coefficient of variation around the grand mean from a single source. In the present study such a partial coefficient of variation CV_p is designated the variability of the effect in question:

$$s^2_{p(i)} = \frac{SSD_{(i)}}{f_t} \qquad \text{(Eq.B.12)}$$

where $s^2_{p(i)}$ is the partial variance from source (i) and

$$CV_{p(i)} = (e^{s^2_{p(i)}} - 1)^{\frac{1}{2}}. \qquad \text{(Eq.B.13)}$$

The radioecological variability depended upon four variables: radionuclide, time, sample species and location. A comparison of, e.g., the time variability among radionuclides assumed sample species, area and period identical for the various nuclides. However, minor differences did not necessarily invalidate the comparison, e.g. whether samples came from 1963-1974, 1962-1974 or 1962-1973 did not influence the variabilities significantly. A significance test between two variabilities was performed on the partial variances as a v^2-test (cf.B.3).

The advantage of the partial variance as compared to the common variance in the anova table is its relative insensitivity to the number of data in the anova. An example may elucidate this. Let us suppose we have two anovas of identical data materials, one with single determinations only and the other one with identical double determinations (error variance = 0). In this case the variance of a main effect would be twice as high in the second anova as in the first, while the partial variances in the two anovas would be the same.

B.5. Anova and variability tables

The tables in the following are numbered B followed by three digits. The two first digits denote, respectively, chapter and section in the main text, and the last is a serial number.

The anova tables shown are a selection of the more than one thousand anovas performed on the data. Special emphasis has been attached to the cases of significant interactions, which, as mentioned above, create problems in the significance tests.

The tables with variabilities display the partial coefficients of variations (CV_p). The partial variance (s_p^2) may be obtained from:

$$s_p^2 = \ln (CV_p^2 + 1) \qquad \text{(Eq.B.14)}$$

Table B.1.2.1. Anova of ln (fCi ^{90}Sr m^{-3}) for ground level air collected in 1957-1975 at Risø (cf. Table A.1.1.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	393.215	18	21.845	307.7	(***)
	Month (M)	99.473	11	9.043	127.4	(***)
Interaction	Y×M	50.979	183	0.279	3.92	***
Replication	Error	9.585	135	0.071		

Although the interaction between years and months was highly significant, the main factors were also highly significant.

Table B.1.2.2. Anova of ln (fCi ^{137}Cs m^{-3}) for ground level air collected in 1957-1975 at Risø

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	317.009	18	17.612	146.77	(***)
	Month (M)	82.173	11	7.470	62.25	(***)
Interaction	Y×M	42.025	188	0.224	1.87	*
Replication	Error	2.873	24	0.120		

The main factors were both highly significant, although the interaction was probably significant.

Table B.1.2.3. The variability of ^{90}Sr and ^{137}Cs in ground level air

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$
Air[a]	Risø	1957-75	fCi ^{90}Sr m^{-3}	347	1.45	0.58
Air[c]	- " -	- " -	fCi ^{137}Cs m^{-3}	241	1.65	0.64
Air[e]	- " -	1962-75	fCi ^{90}Sr and ^{137}Cs m^{-3}	484	1.72	0.61
Air[b]	- " -	1962-72	fCi ^{90}Sr m^{-3}	266	1.48	0.63
Air[b]	- " -	1962-74	- " -	333	1.59	0.61
Air[b]	- " -	1962-76	- " -	405	1.81	0.60
Air[d]	- " -	1962-72	fCi ^{137}Cs m^{-3}	192	1.51	0.62
Air[d]	- " -	1962-74	- " -	216	1.72	0.63
Air[d]	- " -	1962-76	- " -	240	1.88	0.62

a) These air samples consisted of paper filters from the small air-sampler and glass-fibre filters from the large one (Table A.1.1.2).

b) The samples were identical to a), but since 1971 2 separate glass-fibre filters were analyzed each month.

c) Double samples in 1966-1967.

d) Double samples in 1966-1971.

e) Double ^{90}Sr determinations in 1964-1975 (except 1965) and double ^{137}Cs determinations in 1966-1967.

Table B.1.3.1. Anova of ln (pCi ^{90}Sr l^{-1}) precipitation collected in 1962-1974 at Danish state experimental farms (cf. Table A.1.1.3.1)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	1129.458	12	94.121	1448.02	(***)
	Month[a] (M)	233.780	5	46.756	719.32	(***)
	Location (L)	6.176	9	0.686	10.55	(*)
First-order interactions	Y×M	65.258	59	1.106	17.02	***
	Y×L	13.775	108	0.128	1.97	***
	M×L	3.341	45	0.074	1.14	
Second-order interaction	Y×M×L	33.616	519	0.065		

Anovas of the individual years showed that the variations among months were highly significant. The local variation was only significant in 1968-1971 when the stations on Funen and Lolland-Falster showed lower concentrations than Bornholm and Jutland. The variation among years was highly significant, despite the significant interaction between years and months and between years and locations.

As there were no replicates, the second-order interaction was used as error.

a) Bimonthly samples.

Table B.1.3.2. Anova of ln (mCi ^{90}Sr km^{-2} (2 months)$^{-1}$) based on precipitation samples collected in 1962-1974 at Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	1052.744	12	87.729	1235.62	(***)
	Month[a] (M)	208.394	5	41.679	587.03	(***)
	Location (L)	18.674	9	2.075	29.23	(***)
First-order interactions	Y×M	101.992	59	1.729	24.35	***
	Y×L	13.155	108	0.122	1.72	***
	M×L	5.298	45	0.118	1.66	**
Second-order interaction	Y×M×L	36.799	520	0.071		

In contrast to pCi l^{-1} deposition showed a highly significant local variation within the individual years and so did the variation among months. The variation among years was highly significant.

a) Bimonthly samples.

Table B.1.3.3. Anova of ln (mCi ^{90}Sr km^{-2} y^{-1}) based on samples collected in 1960-1974 at Danish state experimental farms and Risø, and at 2 Faroese and 5 Greenlandic stations (cf. Tables A.1.1.3.1 and A.1.1.3.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	316.834	14	22.631	404.12	***
	Location (L)	55.278	18	3.071	54.84	***
Interaction	Y×L	14.896	196	0.076	1.36	
Replication[a]	Error	0.555	10	0.056		

a) The two sampling systems at Risø: rain-bottles and ion-exchange columns were used for the error estimate.

Table B.1.3.4. The variability of ^{90}Sr in precipitation samples

Area	Period	Nuclide unit[c]	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Denmark	1962-74	mCi ^{90}Sr km^{-2} bi.$^{-1}$	758	1.73	0.56[a]	0.16
Faroes	1963-74	mCi ^{90}Sr km^{-2} mo.$^{-1}$	277	1.68	0.19	0.32
Greenland	1963-74	mCi ^{90}Sr km^{-2} qa.$^{-1}$	177	1.70	0.26[b]	0.73
Denmark	1962-74	pCi ^{90}Sr l^{-1}	757	1.68	0.60[a]	0.09
Faroes	- " -	- " -	290	1.78	0.38	0.01
Greenland	- " -	- " -	180	1.75	0.35[b]	0.23
Risø	1962-72	mCi ^{90}Sr km^{-2} mo.$^{-1}$	295	1.36	0.68	-
- " -	- " -	pCi ^{90}Sr l^{-1}	294	1.29	0.63	-

a) Bimonthly samples (Table A.1.1.3.1).

b) Quarterly samples (Table A.1.1.3.2).

c) mo.: month, bi.: 2 months, qa.: 3 months

Table B.1.4.1. The variability of ^{90}Sr in samples of fresh water (cf. Tables A.1.1.4.1 and A.1.1.4.2)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Ground water[a)]	Denmark	1961-74	fCi ^{90}Sr l^{-1}	105	0.57	-	0.49
Stream water	- " -	1971-75	pCi ^{90}Sr l^{-1}	37	-	0.12[b)]	0.43
Lake water	- " -	1971-74	pCi ^{90}Sr l^{-1}	36	-	0.09[b)]	0.83
Drinking water	- " -	1965-71	fCi ^{90}Sr l^{-1}	103	0.65	-	0.79
- " -	Faroes	1962-74	pCi ^{90}Sr l^{-1}	252	0.55	0.12[c)]	0.27
- " -	Greenland	1962-74	pCi ^{90}Sr l^{-1}	170	0.61	-	1.14

a) All locations (cf. A) except Feldbak.

b) The samples were collected in March 1971, September 1971, March 1973, March 1975 and September 1975.

c) The samples were collected every second month: January, March, etc.

Table B.1.5.1. The variability of ^{90}Sr and ^{137}Cs in seawater samples (cf. Table A.1.1.5)

Sample	Salinity	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(locations)}$
"Baltic water"	$\leq 10^{o}/oo$	Inner Danish waters	1961-74	pCi ^{90}Sr l^{-1}	71	0.26	0.05
"North Sea water"	$\geq 33^{o}/oo$	Inner Danish waters, North Sea	1963-74	- " -	44	0.18	0.10
Atlantic surface water	$\sim 34^{o}/oo$	North Atlantic Ocean	1962-74	- " -	44	0.48	0.04
Surface water	$\sim 29^{o}/oo$	Greenland waters	1962-74	- " -	67	0.46	0.08
"Baltic water"	$\leq 10^{o}/oo$	Inner Danish waters	1972-74	pCi ^{137}Cs l^{-1}	28	0.02	0.05
"North Sea water"	$\geq 33^{o}/oo$	Inner Danish waters, North Sea	- " -	- " -	38	0.20	0.07

Table B.1.6.1. The variability of ^{90}Sr in Danish soil (cf. Table A.1.1.6.1)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(locations)}$
0-20 cm layer	Denmark	1962-71[a)]	mCi ^{90}Sr km^{-2}	206	0.18	0.13
Total depth[e)]	- " -	1962-74	- " -	128[f)]	0.20	0.13
0-30 cm	Jutland[b)]	1971	- " -	20	-	0.19[d)]
- " -	Islands[c)]	- " -	- " -	26	-	0.43[d)]

a) Except 1970, March 1963 was used as 1962 (Fig. 1.6.1)

b) Tylstrup, Studsgård, Jyndevad (sandy soils).

c) Blangstedgård, Tystofte, Virumgård, Åkirkeby (loamy soils).

d) Variability among depths: 0-10, 10-20 and 20-30 cm.

e) 1962-1969: 0-20 cm, 1970-1973: 0-30 cm, 1974-1975: 0-50 cm.

f) The anova was performed on the mean values only.

Table B.2.2.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in cereal grain collected in 1959-1974 at the Danish state experimental farms (cf. Table A.1.2.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	565.560	15	37.704	942.60	(***)
	Species (S)	77.286	3	25.762	644.05	(***)
	Location (L)	158.720	10	15.872	396.80	(***)
First-order interactions	Y×S	17.010	45	0.378	9.45	(***)
	Y×L	49.545	135	0.367	9.18	(***)
	S×L	7.950	30	0.265	6.63	(***)
Second-order interaction	Y×S×L	36.668	356	0.103	2.58	***
Replication	Error	22.520	563	0.040		

As the second-order interaction was highly significant, the test of first-order interactions and main factors had to be performed in two-way and one-way anovas. This breakdown analysis, however, showed that both first-order interactions as well as main factors were significant in most cases.

Table B.2.2.2. Anova of ln (pCi ^{90}Sr kg^{-1}) in cereal grain collected in 1959-1974 at the Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	627.225	15	41.815	1306.72	(***)
	Species (S)	3.225	3	1.075	33.59	
	Location (L)	136.420	10	13.642	426.31	(***)
First-order interactions	Y×S	28.620	45	0.636	19.88	(***)
	Y×L	55.620	135	0.412	12.88	(***)
	S×L	6.120	30	0.204	6.38	(***)
Second-order interaction	Y×S×L	39.050	355	0.110	3.44	***
Replication	Error	18.274	564	0.032		

Cf. the remarks to Table B.2.2.1; however, regarding the variation among species, 1965 did not show any significant difference because in that year the preferential direct contamination of rye was just outbalanced by the preferential indirect contamination of wheat and oats.

Table B.2.2.3. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in cereal grain collected in 1962-1974 at the Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	950.880	12	79.240	1495.09	(***)
	Species (S)	18.300	3	6.100	115.09	(***)
	Location (L)	21.020	10	2.102	39.66	(***)
First-order interactions	Y×S	4.716	36	0.131	2.47	
	Y×L	17.538	111	0.158	2.98	
	S×L	3.390	30	0.113	2.13	
Second-order interaction	Y×S×L	25.844	284	0.091	1.72	***
Replication	Error	5.872	111	0.053		

Break-down analysis showed that the first-order interactions were not significant, while the main factors were all highly significant.

Table B.2.2.4. Anova of ln (pCi ^{137}Cs kg^{-1}) in cereal grain collected in 1962-1974 at the Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	849.264	12	70.772	1228.73	(***)
	Species (S)	22.140	3	7.380	128.13	(***)
	Location (L)	21.250	10	2.125	36.89	(***)
First-order interactions	Y×S	4.320	36	0.120	2.08	
	Y×L	16.650	111	0.150	2.60	
	S×L	4.350	30	0.145	2.52	
Second-order interaction	Y×S×L	26.790	285	0.094	1.63	***
Replication	Error	6.336	110	0.058		

Cf. remarks to Table B.2.2.3.

Table B.2.2.5. Anova of ln (pCi ^{54}Mn kg^{-1}) cereal grain collected in 1962-1965 at the Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	258.100	2	129.050	941.97	•••
	Species (S)	1.660	3	0.553	4.04	••
	Location (L)	8.950	9	0.994	7.25	•••
First-order interactions	Y×S	1.075	6	0.179	1.31	
	Y×L	2.594	18	0.144	1.05	
	S×L	3.083	27	0.114	0.83	
Second-order interaction	Y×S×L	6.010	44	0.137	1.00	
Replication	Error	2.869	21	0.137		

Table B.2.2.6. Anova of ln (mg Sr (g Ca)$^{-1}$) in cereal grain collected in 1962-1971 at the Danish state experimental farms

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	1.360	9	0.151	2.32	
	Species (S)	38.831	3	12.944	198.53	(•••)
	Location (L)	15.771	10	1.577	24.19	(•••)
First-order interactions	Y×S	3.156	27	0.117	1.80	•
	Y×L	5.706	77	0.074	1.14	
	S×L	3.930	30	0.131	2.01	••
Second-order interaction	Y×S×L	11.492	170	0.068	1.04	
Replication	Error	4.955	76	0.065		

Two-way anovas of the individual years and species showed highly significant variations among species and among locations.

The samples from 1962-1971 were all re-analysed in 1972. The anova was performed on the results from this re-analysis.

Table 8.2.2.7. The variability of ^{90}Sr concentrations in Danish cereal grain (cf. Table A.1.2.2)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
All species	Denmark[a]	1959-74	pCi ^{90}Sr kg^{-1}	1157	0.85	0.05	0.35
- " -	- " -	1962-66	- " -	421	0.68	0.09	0.29
- " -	- " -	1967-71	- " -	389	0.24	0.11	0.42
- " -	Jutland[b]	1962-66	- " -	158	0.69	0.12	0.09
- " -	- " -	1967-72	- " -	186	0.21	0.16	0.19
- " -	Islands[c]	1962-66	- " -	184	0.68	0.24	0.07
- " -	- " -	1967-72	- " -	235	0.28	0.11	0.14
Rye	Denmark[a]	1959-73	pCi ^{90}Sr (g Ca)$^{-1}$	232	1.01	-	0.28
Barley	- " -	- " -	- " -	264	0.88	-	0.43
Wheat	- " -	- " -	- " -	361	0.70	-	0.40
Oats	- " -	- " -	- " -	255	0.72	-	0.42
Rye	Jutland[b]	- " -	- " -	115	1.02	-	0.10
Barley	- " -	- " -	- " -	102	0.98	-	0.20
Wheat	- " -	- " -	- " -	110	0.58	-	0.21
Oats	- " -	- " -	- " -	100	0.73	-	0.19
Rye	Islands[c]	- " -	- " -	80	0.99	-	0.12
Barley	- " -	- " -	- " -	124	0.84	-	0.13
Wheat	- " -	- " -	- " -	193	0.79	-	0.19
Oats	- " -	- " -	- " -	119	0.72	-	0.18

a) The eleven state experimental farms.

b) The farms in Jutland except Ødum.

c) The farms on the Islands except Virumgård.

Table 8.2.2.8. The variability of ^{137}Cs concentrations in Danish cereal grain (cf. Table A.1.2.2)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
All species	Denmark[a]	1962-74	pCi ^{137}Cs kg^{-1}	597	1.77	0.19	0.19
- " -	- " -	1962-66	- " -	223	0.95	0.21	0.20
- " -	- " -	1967-71	- " -	243	0.35	0.21	0.19
- " -	Jutland[b]	1962-66	- " -	82	1.05	0.23	0.16
- " -	- " -	1967-72	- " -	110	0.44	0.23	0.15
- " -	Islands[c]	1962-66	- " -	96	0.94	0.21	0.05
- " -	- " -	1967-72	- " -	144	0.45	0.18	0.11
Rye	Denmark[a]	1962-72	- " -	108	1.58	-	0.21
Barley	- " -	- " -	- " -	123	1.84	-	0.18
Wheat	- " -	- " -	- " -	169	1.44	-	0.22
Oats	- " -	- " -	- " -	113	1.52	-	0.25
Rye	Jutland[b]	- " -	- " -	53	1.77	-	0.18
Barley	- " -	- " -	- " -	45	2.09	-	0.13
Wheat	- " -	- " -	- " -	50	1.45	-	0.15
Oats	- " -	- " -	- " -	42	1.54	-	0.18
Rye	Islands[c]	- " -	- " -	35	1.49	-	0.05
Barley	- " -	- " -	- " -	58	1.64	-	0.09
Wheat	- " -	- " -	- " -	90	1.45	-	0.05
Oats	- " -	- " -	- " -	53	1.47	-	0.22

a) The eleven state experimental farms.

b) The farms in Jutland except Ødum.

c) The farms on the Islands except Virumgård.

Table B.2.2.9. The variability of various radionuclides and stable elements in all species of Danish cereal grain

Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
Denmark	1963-65	pCi ^{54}Mn kg^{-1}	130	-	0.11	0.26
- " -	1963 and 1965	pCi $^{239,240}Pu$ kg^{-1}	91	-	0.73	0.21
- " -	1962 and 1963	pCi ^{89}Sr kg^{-1}	110	-	0.09	0.29[a]
- " -	1962-71	mg Sr (g Ca)$^{-1}$	402	0.06	0.32	0.20
- " -	1962-72	g Ca kg^{-1}	883	0.06	0.28	0.06
- " -	1962-73	g K kg^{-1}	550	0.09	0.07	0.03

a) The local variability was enhanced due to a contribution from time variability because the date of harvesting differed at the various locations. This influences the levels found of a short-lived nuclide such as ^{89}Sr.

Table B.2.3.1. The variability of radionuclide concentrations in bread (cf. Table A.1.2.3)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
Rye bread	Denmark	1962-74	pCi ^{90}Sr kg^{-1}	334	1.03	-	0.13
- " -	- " -	1963-74	pCi ^{137}Cs kg^{-1}	169	1.93	-	0.12
White bread	- " -	1962-74	pCi ^{90}Sr kg^{-1}	297	0.76	-	0.04
- " -	- " -	1963-74	pCi ^{137}Cs kg^{-1}	173	2.12	-	0.06
Rye and white bread	- " -	1963-65	pCi ^{54}Mn kg^{-1}	72	0.80	0.94	0.18
- " -	Faroes	1964-72	pCi ^{90}Sr kg^{-1}	41	0.70	0.87	-
- " -	- " -	1964-72	pCi ^{137}Cs kg^{-1}	32	2.00	0.41	-

a) Samples were measured each June and December.

Table B.2.4.1. The variability of ^{90}Sr and ^{137}Cs in grass (cf. Table A.1.2.4)

Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Denmark[a]	1962-70	pCi ^{90}Sr (g Ca)$^{-1}$	116	1.00	-	0.22
W.Greenland[b]	1962-67	- " -	43	0.38	0.04	0.53
Faroes	1962-75	- " -	31	0.85	0.08	-
Denmark[a]	1964-70	pCi ^{137}Cs (g K)$^{-1}$	54	1.11	-	0.35
W.Greenland[b]	1964-67	- " -	22	0.32	0.23	0.32
Faroes	1964-72	- " -	19	0.64	0.15	-

a) The state experimental farms.

b) Julianehåb, Godthåb, Sukkertoppen, Holsteinsborg and Egedesminde.

Table B.2.5.1. Anova of ln (pCi $^{90}Sr\ kg^{-1}$) fresh weight in Danish white cabbage collected in 1962-1974 in the 8 zones (cf. Table A.1.2.5.1)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	4.795	11	0.436	3.61	***
	Location (L)	4.054	7	0.579	4.80	***
Interaction	Y×L	11.356	70	0.162	1.34	
Replication	Error	6.513	54	0.121		

Table B.2.5.2. Anova of ln (pCi $^{90}Sr\ kg^{-1}$) fresh weight in Danish potatoes collected in 1962-1974 at the ten Danish state experimental farms (cf. Table A.1.2.5.1)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	4.879	12	0.407	11.63	(***)
	Location (L)	5.767	10	0.577	16.49	(***)
Interaction	Y×L	17.854	105	0.170	4.79	***
Replication	Error	3.016	85	0.035		

The interaction between years and locations was highly significant, but the general trend of the main factors was evident: the maximum occurred in 1963-1964 and the lowest levels were found in 1962 and in 1974, although a few farms deviated from this pattern. Studsgård, Askov and Jyndevad generally showed the highest levels, although deviations occurred in certain years.

Table B.2.5.3. The variability of ^{90}Sr, stable Sr and ^{137}Cs in vegetables and fruits

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
Cabbage	Denmark	1962-74	pCi $^{90}Sr\ kg^{-1}$	142	0.18	-	0.17[a]
Carrots	- " -	1963-74	- " -	109	0.23	-	0.33[a]
Potatoes	- " -	1962-74	- " -	212	0.15	-	0.17[b]
Cabbage	- " -	1963-74	pCi $^{137}Cs\ kg^{-1}$	11	0.99[e]	-	-
Carrots	- " -	1963-74	- " -	11	1.58[e]	-	-
Potatoes	- " -	1963-74	- " -	11	1.13[e]	-	-
Apples	- " -	1962-74	pCi $^{90}Sr\ kg^{-1}$	93	0.97	-	0.10[a]
- " -	- " -	1963-74	pCi $^{137}Cs\ kg^{-1}$	11	1.32[e]	-	-
"Greens"	- " -	1960-66	pCi $^{90}Sr\ kg^{-1}$	267	0.27	0.88	0.15[a]
"Roots"	- " -	- " -	- " -	270	0.31	0.89	0.18[a]
Berries	- " -	- " -	- " -	58	0.33	0.79	0.23[a]
"Greens"	- " -	1963-66	pCi $^{137}Cs\ kg^{-1}$	152	0.64	0.83	0.11[a]
"Roots"	- " -	- " -	- " -	110	0.73	0.55	0.24[a]
Berries	- " -	- " -	- " -	41	0.44	0.49	0.24[a]
"Greens"	- " -	1960-66	mg Sr $(g\ Ca)^{-1}$	59	0.12	0.28	0.17[a]
"Roots"	- " -	- " -	- " -	50	0.12	0.22	0.18[a]
Leaves brussel sprouts	- " -	1962-63	pCi $^{90}Sr\ kg^{-1}$	95	0.24	0.10[d]	0.31[c]
- " -	- " -	1963	pCi $^{137}Cs\ kg^{-1}$	21	-	0.24[d]	0.32[c]
- " -	- " -	1962-63	mg Sr $(g\ Ca)^{-1}$	68	0.02	0.25[d]	0.43[c]

a) The 8 zones (Table A.1.2.5.1).
b) The 10 state experimental farms (Table A.1.2.5.1).
c) The 6 growing stations (Table A.1.2.5.1).
d) $CV_{p(months)}$
e) Each year was represented by one value only:
"Greens": white cabbage, spring cabbage, red cabbage, brussel sprouts, kale, cauliflower, cucumber, peas, beans, lettuce, spinach, parsley.
"Roots": potatoes, carrots, onion, leek, beetroot, celeriac.
Berries: strawberry, tomato, gooseberry.

Table B.2.5.4. Anova of ln (pCi $^{90}Sr\ kg^{-1}$) apples collected in 1962-1974 in the 8 zones (cf. Table A.1.2.5.1)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	62.011	12	5.168	28.9	***
	Location (L)	0.918	7	0.131	0.73	
Interaction	Y×L	11.151	69	0.162	0.90	
Replicatien	Error	0.893	5	0.179		

Table B.2.6.1. The variability of ^{90}Sr and ^{137}Cs in West Greenland lichen (cf. Table A.1.2.6)

Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(locations)}$
W.Greenland	1962-75	pCi ^{90}Sr kg^{-1}	32	0.41	0.05
- " -	- " -	pCi ^{137}Cs kg^{-1}	31	0.27	0.14

Table B.2.7.1. The variability of ^{90}Sr in sea plants (cf. Table A.1.2.7)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$
Fucus vesiculosus	Zealand	1960-75	pCi ^{90}Sr (g Ca)$^{-1}$	30	0.63
Zostera marina	- " -	- " -	- " -	34	0.65

Table B.3.2.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in Danish dried milk collected monthly from May 1959 to April 1976 ("milk year") from the seven dried-milk factories (cf. Table A.1.3.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	656.176	16	41.011	2278.39	(***)
	Month (M)	2.926	11	0.266	14.78	
	Location (L)	118.884	6	19.814	1100.78	(***)
First-order interactions	Y×M	17.716	172	0.103	5.72	(***)
	Y×L	6.552	91	0.072	4.00	(***)
	M×L	3.630	66	0.055	3.06	(***)
Second-order interaction	Y×M×L	25.984	928	0.028	1.56	***
Replication	Error	8.101	438	0.018		

As the second-order interaction is highly significant, it is necessary to make a "breakdown analysis", *e.g. consider one location at a time and one year.* Such a breakdown analysis reveals a highly significant variation among years and among locations, while the monthly variation is less evident due to the significant interaction between years and months.

Table B.3.2.2. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in Danish dried milk collected monthly from May 1957 to April 1976 ("milk year") from the seven dried milk factories (cf. Table A.1.3.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	1183.230	18	65.735	1195.18	(***)
	Month (M)	117.351	11	10.641	193.47	(***)
	Location (L)	178.902	6	29.817	542.13	(***)
First-order interactions	Y·M	37.714	173	0.218	3.96	***
	Y·L	17.472	91	0.192	3.49	***
	M·L	7.194	66	0.109	1.98	**
Second-order interaction	Y·M·L	79.390	934	0.085	1.55	*
Replication	Error	2.957	54	0.055		

The second-order interaction is only probably significant and may thus be neglected. The first-order interactions are, however, all significant, and a breakdown analysis is necessary. This analysis indicates a highly significant variation among years, months and locations.

Table B.3.2.3. The variability of concentrations of ^{90}Sr and ^{137}Cs in Danish milk ("milk year") (cf. Table A.1.3.2)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Dried milk	7 factories	1959-75	pCi ^{90}Sr (g Ca)$^{-1}$	1728	0.68	0.04	0.26
- " -	- " -	1962-76	- " -	1226	0.70	0.04	0.28
- " -	- " -	1961-72	- " -	1292	0.62	0.04	0.26
Fresh milk	8 zones	1962-76	- " -	239	0.64	0.00[a]	0.22
Whole milk	9 exp.farms	1962-70	- " -	84	0.52[b]	-	0.21
Dried milk	7 factories	1957-75	pCi ^{137}Cs (g K)$^{-1}$	1353	1.18	0.30	0.38
- " -	- " -	1962-76	- " -	1230	1.45	0.29	0.39
- " -	- " -	1961-72	- " -	926	1.02	0.34	0.39
Fresh milk	8 zones	1962-76	- " -	237	1.53	0.19[a]	0.27
Whole milk	9 exp.farms	1962-70	- " -	73	1.26[b]	-	0.26

a) Only June and December were included for fresh milk in the monthly variability.

b) The whole milk samples were collected in September only.

Table B.3.3.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in Faroese fresh milk collected weekly and analysed monthly; May 1962 - April 1976 ("milk year"), from Thorshavn, Klaksvig and Tvørå (cf. Table A.1.3.3)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	236.340	13	18.180	649.29	(***)
	Month (M)	8.096	11	0.736	26.29	
	Location (L)	1.622	2	0.811	28.96	
First-order interactions	Y×M	12.922	142	0.091	3.25	***
	Y×L	2.834	26	0.109	3.89	***
	M×L	1.166	22	0.053	1.89	
Second-order interaction	Y×M×L	10.080	280	0.036	1.29	
Replication	Error	1.722	61	0.028		

Two of the first-order interactions were highly significant. Individual anovas for the years showed that neither the variation among locations nor the variation among months was generally significant; but the variation among years was highly significant within each location.

Table B.3.3.2. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in Faroese fresh milk collected weekly and analysed monthly; May 1962 - April 1976 ("milk year"), from Thorshavn, Klaksvig and Tvørå

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	185.406	13	14.262	178.28	(***)
	Month (M)	4.411	11	0.401	5.01	
	Location (L)	41.362	2	20.681	258.51	(***)
First-order interactions	Y×M	19.734	143	0.138	1.73	***
	Y×L	5.252	26	0.202	2.53	***
	M×L	4.796	22	0.218	2.73	***
Second-order interaction	Y×M×L	22.160	277	0.080	-	
Replication	Error	-	-	-	-	

The first-order interactions were highly significant. Individual anovas for the years showed that the variation among locations was highly significant, whereas the variation among months was generally insignificant. The variation among years was highly significant within each location.

Table B.3.3.3. The variability of concentrations of ^{90}Sr and ^{137}Cs in Faroese milk ("milk year") (cf. Table A.1.3.3)

Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Faroes	1962-76	pCi ^{90}Sr (g Ca)$^{-1}$	557	0.73	0.12	0.65
- " -	- " -	pCi ^{137}Cs (g K)$^{-1}$	494	0.67	0.09	0.29
Thorshavn	- " -	- " -	117	0.60	0.19	-
Klaksvig	- " -	- " -	118	0.54	0.10	-
Tværå	- " -	- " -	114	0.43	0.10	-
Faroes	1962-64	- " -	65	0.40	0.20	0.17
- " -	1964-67	- " -	106	0.34	0.12	0.23
- " -	1967-72	- " -	178	0.13	0.16	0.31
- " -	1972-76	- " -	143	0.14	0.11	0.40

Table B.3.4.1. The variability of concentrations of ^{137}Cs and ^{90}Sr in terrestrial animals (cf. Tables A.1.3.4.1 and A.1.3.4.2)

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$	$CV_{p(locations)}$
Beef and veal	Copenhagen	June 63-March 76	pCi ^{137}Cs kg^{-1}	69	1.16	0.24[a]	- [b]
Pork	- " -	- " -	- " -	47	1.57	0.09[a]	- [b]
Beef and veal	- " -	- " -	pCi ^{90}Sr kg^{-1}	64	0.60	0.17[a]	-
Pork	- " -	- " -	- " -	44	0.82	0.13[a]	-
Reindeer	W.Greenland	1962-74	pCi ^{137}Cs kg^{-1}	43	1.62	-	0.36[c]
- " -	- " -	1962-72	- " -	42	1.19	0.57[f]	-
- " -	- " -	1962-74	pCi ^{90}Sr kg^{-1}	42	1.10	-	0.10[c]
- " -	- " -	1962-72	- " -	41	0.99	0.18[f]	-
Reindeer bone	- " -	1962-75	pCi ^{90}Sr (g Ca)$^{-1}$	53	0.88	-	0.19[d]
Mutton	- " -	1961-75[e]	pCi ^{137}Cs kg^{-1}	48	0.99	-	-
- " -	Faroes	1962-76	- " -	43	0.79	-	-
Sheep bone	- " -	1962-76	pCi ^{90}Sr (g Ca)$^{-1}$	40	0.74	-	-

a) The samples were collected in June, September, December, and March.

b) In December 1963, meat was sampled in the 8 zones; $CV_{p(locations)}$ were on that occasion determined at 0.33 for beef and 0.30 for pork (^{137}Cs).

c) 3 locations were included for reindeer meat sampling.

d) 5 locations were included for reindeer bone sampling.

e) No samples of Greenland mutton were available in 1963, 1964, 1966, 1967 and 1970.

f) The samples were from late winter and autumn (two periods).

Table B.3.5.1. The variability of concentrations of ^{137}Cs and ^{90}Sr in fish (cf. Table A.1.3.5)

Sample[b]	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$
8 species	Denmark[a]	1963-73	pCi ^{137}Cs kg^{-1}	60	0.26	0.50
2 - " -	Faroes	1962-76	- " -	95	0.40	0.11
5 - " -	Greenland	1963-72	- " -	43	0.46	0.38
8 - " -	Denmark[a]	1963-73	pCi ^{90}Sr kg^{-1}	49	0.31	0.40
2 - " -	Faroes	1962-76	- " -	96	0.56	0.10
5 - " -	Greenland	1963-72	- " -	39	0.53	0.60

a) The Danish fish were caught in inner Danish waters.

b) The Danish species were: cod, plaice, herring, eel, salmon, garfish, mackerel, trout; the Faroese species were: cod and haddock; the Greenland species were: cod, halibut, salmon, angmasatter, trout.

Table B.3.6.1. The variability of ^{90}Sr and ^{137}Cs in seals, whales and hens' eggs

Sample	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$
Whale	N.Atlantic[b]	1962-75[a]	pCi ^{90}Sr kg^{-1}	19	0.81[f]
Seal	Greenland	1962-76[c]	- " -	33	0.98[f]
Whale	N.Atlantic[b]	1962-75[d]	pCi ^{137}Cs kg^{-1}	15	0.89[f]
Seal	Greenland	1962-76[c]	- " -	36	0.44[f]
Hens' eggs	Denmark	1963-75[e]	pCi ^{90}Sr kg^{-1}	10	1.25
- " -	- " -	- " -	pCi ^{137}Cs kg^{-1}	10	1.39

a) Except 1964, 1967, 1971, and 1973.

b) The samples were from the Faroe Islands and from Greenland (Table A.1.3.6.1).

c) Except 1970-1975.

d) Except 1964, 1967 and 1971-1973.

e) Except 1967, 1972 and 1973.

f) The samples were collected over a large area, consequently the variability among years also comprise local variability to some extent. Furthermore the variability among years also contains a contribution from interspecific variability; the Faroese whales were all pilot whales (Globicephala melaena), which feed mostly on cuttlefish, and the Greenland whales were piked whales (Balaenoptera acutorostrata), which feed on fish (especially herring).

Table B.3.6.2. Anova of ^{90}Sr and ^{137}Cs in Danish and Faroese eggs collected in 1963-1976 (unit: ln pCi kg^{-1})

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	40.468	13	3.113	8.67	***
	Location (L)	1.609	1	1.609	4.48	*
	Nuclide (N)	20.549	1	20.549	57.24	***
First-order interactions	Y×L	5.916	8	0.739	1.13	
	Y×N	1.734	12	0.145	0.22	
	L×N	0.072	1	0.072	0.11	
Second-order interaction	Y×L×N	2.689	8	0.336	0.51	
Replication	Error	15.058	23	0.655		

The Faroese eggs probably contained more ^{90}Sr and ^{137}Cs than the Danish (1.4 times more on the average). The concentration of ^{137}Cs was 3 times that of ^{90}Sr. The replication includes variations within a sampling year and may thus also include variations due to the different origins of the eggs.

Table B.4.2.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in Danish total diet collected in 1962-1974 in the 8 zones in June and December (cf. Table A.1.4.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Time (T)	141.125	25	5.645	217.1	***
	Location (L)	9.137	7	1.331	51.2	***
Interaction	T×L	5.600	175	0.032	1.23	*
Replication	Error	6.502	254	0.026		

Time means six months; the interaction between time and location was only probably significant. It is therefore neglected in the tests of the main factors, which are both highly significant.

Table B.4.2.2. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in Danish total diet collected in 1963-1974 in the 8 zones in June and December (cf. Table A.1.4.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Time (T)	361.813	23	15.731	(491.6)	(***)
	Location (L)	5.950	7	0.850	(26.6)	(***)
Interaction	T×L	11.592	161	0.072	2.25	***
Replication	Error	5.735	177	0.032		

Time means six months. The interaction is highly significant, and the test of the main factors may thus be unwarranted. However, an inspection of the data (Fig. 4.2.1) shows a substantial overall effect of T, and until 1972 it is further evident that the local variation is significant (West-Jutland > Lolland-Falster).

Table B.4.2.3. The variability in Danish total diet samples (cf. Table A.1.4.2)

Area	Period	Nuclide unit	f_t	$CV_{p(years)'}$	$CV_{p(locations)}$
8 zones	1962-74	pCi ^{90}Sr (g Ca)$^{-1}$	461	0.60	0.14
- " -	1961-66	- " -	264	0.67	0.13
- " -	1967-72	- " -	191	0.14	0.16
- " -	1973-76	- " -	127	0.32	0.10
8 zones	1963-74	pCi ^{137}Cs (g K)$^{-1}$	368	1.29	0.13
- " -	1963-66	- " -	126	0.52	0.16
- " -	1967-72	- " -	205	0.22	0.12
- " -	1973-76	- " -	67	0.28	0.12

(years)' means periods of six months (cf. Table B.4.2.1 and Table B.4.2.2). The local variability showed a decreasing tendency with time reflecting the increasing transfer of foodstuffs among the various areas of the country. The local variability may be even less pronounced than seen in 1973-1976 because the number of towns within the sampling zones in this period was reduced from 6 to 1.
If the yearly diet activity levels were considered (i.e. 1/4 December diet (i-1) + 1/2 June diet (i) + 1/4 December diet (i)) $CV_{p(years)}$ became 0.59 for ^{90}Sr (1962-1974) and 1.48 for ^{137}Cs)1963-1974).

Table B.4.2.4. Anova of ln (mg Sr (g Ca)$^{-1}$) in Danish total diet collected in 1963-1972 in the 8 zones in June and December (cf. Table A.1.4.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	0.939	9	0.104	2.60	**
	Location (L)	21.700	8	2.712	67.80	***
	Month (M)	1.431	1	1.431	35.78	***
First-order interactions	Y×L	3.164	71	0.045	1.12	
	Y×M	0.642	8	0.080	2.00	
	L×M	0 110	7	0.016	0.40	
Second-order interaction	Y×L×M	1.427	49	0.029	0.73	
Replication	Error	5.199	130	0.040		

The mean of the 8 zones was 1.44 mg Sr (g Ca)$^{-1}$, and the population weighted mean (Copenhagen included) was 1.52 mg Sr (g Ca)$^{-1}$.

Table B.4.2.5. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$)$_{DIET}$/(pCi ^{90}Sr (g Ca)$^{-1}$)$_{MILK}$ collected simultaneously in the 8 zones in Denmark during 1962-1976 (cf. Tables A.1.4.2 and A.1.3.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	23.837	29	0.822	35.7	(***)
	Location (L)	4.501	7	0.643	28.0	(***)
Interaction	Y×L	17.763	203	0.088	3.77	***
Replication	Error	6.645	286	0.023		

The arithmetic grand mean was 1.44. In general, the Islands showed higher ratios than Jutland (1.18 times higher). The maximum ratios occurred in June 1974 and in June 1966. Minima were in December 1970 and December 1967. The ratio between max. and min. was approx. 2. The interaction was partly due to a decreasing difference with time between the ratios found in Jutland and the Islands. The tranfer of rye from Jutland to the Islands and of wheat in the other direction explains the local variation.

Table B.4.2.6. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$)$_{DIET}$/(pCi ^{137}Cs (g K)$^{-1}$)$_{MILK}$ collected simultaneously in the 8 zones in Denmark during 1962-1976 (cf. Tables A.1.4.2 and A.1.3.2)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	37.338	27	1.383	40.7	(***)
	Location (L)	8.457	7	1.208	35.5	(***)
Interaction	Y×L	38.613	187	0.206	6.14	***
Replication	Error	5.917	176	0.034		

The arithmetic grand mean was 1.57. The local pattern was similar to that observed for ^{90}Sr (Table B.4.2.5), but the local variation was more pronounced for ^{137}Cs (Jutland/Islands = 1.27). The time pattern also corresponded to that of ^{90}Sr, but the ratio between max. and min. was approx. 3.

Table B.4.3.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in Danish infant bone collected in 1962-1976 (cf. Table A.1.4.3)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	105.896	14	7.564	33.92	•••
	Age[1] (A)	4.052	4	1.013	4.54	••
	Location[2] (L)	4.616	4	1.154	5.17	••
First-order interactions	Y×A	8.190	39	0.210	0.94	
	Y×L	11.029	41	0.269	1.21	
	A×L	1.836	6	0.306	1.37	
Second-order interaction	Y×A×L	4.675	17	0.275	1.23	
Replication	Error	97.737	438	0.223		

[1] The age groups were 0-1, > 1-2, > 2-3, > 3-4, and > 4-5 years.

[2] The locations were zones I, II, III, IV, V, VI.

Table B.4.3.2. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in Danish adult bone collected in 1960-1976 (cf. Table A.1.4.3)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	96.432	16	6.027	55.29	•••
	Age[1] (A)	2.238	6	0.373	3.42	••
	Location[2] (L)	8.634	6	1.439	13.20	•••
First-order interactions	Y×A	10.370	85	0.122	1.12	
	Y×L	5.050	50	0.101	0.93	
	A×L	2.240	20	0.112	1.03	
Second-order interaction	Y×A×L	11.330	103	0.110	1.01	
Replication	Error	59.732	548	0.109		

[1] The age groups were: 20-29, 30-39, 40-49, 50-59, 60-69, 70-79, ≥ 80 years.

[2] The locations were: zones I-VIII.

Table B.4.3.3. The variability in Danish human bone samples (cf. Table A.1.4.3)

Age	Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(locations)}$
0-30 days	Zones I-VI[a]	1963-76	pCi ^{90}Sr (g Ca)$^{-1}$	167	0.31	0.05
0-5 years	Zones I-VI	1962-76	- " -	563	0.45	0.09
5-11 years	Zones I-VI[a]	1960-76	- " -	151	0.30	0.08
12-19 years	Zones I-VIII	1960-76	- " -	329	0.44	0.06
20-29 years	Zones I-VII	1960-76	- " -	287	0.35	0.11
≥ 30 years	Zones I-VIII	1960-76	- " -	546	0.37	0.09

a) Except zone V.

Table B.4.4.1. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in deciduous teeth shed by Danish children born in 1950-1969 in Jutland and the Islands (cf. Table A.1.4.4)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year of birth (Y)	317.243	19	16.697	181.49	•••
	Location (L)	3.033	1	3.033	32.97	•••
	Species (S)	3.870	3	1.290	14.02	•••
First-order interactions	Y×L	1.972	17	0.116	1.26	
	Y×S	7.850	50	0.157	1.71	••
	L×S	0.144	3	0.048	0.52	
Second-order interaction	Y×L×S	3.198	41	0.078	0.85	
Replication	Error	23.307	254	0.092		

Although the interaction between birth year and the different teeth was significant, this interaction is not so pronounced that it eclipses the significance of the main factors. However, the difference among the different teeth (incisors, cuspids, molar I and molar II) was not significant in all materials (cf. XII).

Table B.4.4.2. The variability in samples of deciduous teeth (cf. Table A.1.4.4)

Area	Period	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(species)}$	$CV_{p(locations)}$
Denmark	1950-69	pCi ^{90}Sr (g Ca)$^{-1}$	388	1.12	0.10	0.09
Faroes	1950-66	- " -	143	1.08	0.08	-
Greenland	1952-67	- " -	70	1.06	0.07	-

Table B.4.5.1.a. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in five adult males working at Risø measured in 1963-1972 (cf. Table A.1.4.5)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	90.355	9	10.039	267.8	•••
	Individual (I)	1.849	4	0.462	12.3	•••
Interaction	Y×I	1.961	33	0.059	1.58	
Replication	Error	2.925	78	0.037		

The three annual measurements were used as replicates, i.e., the error thus includes a possible seasonal variation.

Table B.4.5.1.b. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in five adult females working at Risø measured in 1963-1972

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	113.356	9	12.595	139.7	•••
	Individual (I)	3.397	4	0.849	9.42	•••
Interaction	Y×I	4.345	36	0.128	1.42	
Replication	Error	6.760	75	0.090		

The two groups analysed in Tables a and b were selected from the entire control group according to the criterion that these individuals had been measured regularly throughout the period of observation.

Table B.4.5.2. The variability in wholebody ^{137}Cs concentrations (cf. Table A.1.4.5)

Area	Period	Nuclide unit	f_t	CV_p(years)	CV_p(sexes)	CV_p(individuals)
Risø	1963-72[a)]	pCi ^{137}Cs (g K)$^{-1}$	247	1.13	-	0.20
- " -	1963-69	- " -	192	0.65	0.09	-
- " -	1970-76	- " -	162	0.36	0.20	-

a) The group consisted of the 10 individuals analysed by sex in Tables B.4.5.1.a and b.

Table B.4.6.1.a. Anova of ln (pCi ^{90}Sr (g Ca)$^{-1}$) in human milk collected in 1961-1969 ("milk year") in Zealand (cf. Table A.1.4.6)

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	18.937	8	2.367	27.97	***
	Month (M)	0.934	11	0.085	1.00	
Interaction	Y*M	4.392	28	0.157	1.85	
Replication	Error	2.201	26	0.085		

Table B.4.6.1.b. Anova of ln (pCi ^{137}Cs (g K)$^{-1}$) in human milk collected in 1961-1969 ("milk year") in Zealand

Nature of effect	Source	SSD	f	s^2	v^2	Significance
Main factors	Year (Y)	112.866	8	14.108	(256.5)	(***)
	Month (M)	4.222	11	0.384	(6.98)	(***)
Interaction	Y*M	4.880	34	0.144	2.62	***
Replication	Error	5.655	103	0.055		

Although the interaction was significant, it was evident that the variation among years was highly significant. The mean level in 1963-1964 was an order of magnitude higher than in 1969-1970. The variation among months was also significant. During the entire period the summer months thus showed levels nearly twice as high as those in winter and early spring.

Table B.4.6.2. The variability in Danish human milk samples (cf. Table A.1.4.6)

Area	Period[a]	Nuclide unit	f_t	$CV_{p(years)}$	$CV_{p(months)}$
Zealand	1961-69	pCi ^{90}Sr (g Ca)$^{-1}$	65	0.54	0.11
Zealand	1961-69	pCi ^{137}Cs (g K)$^{-1}$	156	1.03	0.18

a) The years were "milk years", i.e., $May_{(i-1)}$ - $April_{(i)}$.

APPENDIX C

PREDICTION MODELS AND RADIOECOLOGICAL SENSITIVITY

Appendix C describes the calculation of prediction models by the so-called FIT procedure. The models estimated for the various samples studied are given in tables where the radioecological sensitivity is also estimated.

C.1. Introduction

An environmental sample is, in principle, contaminated by radioactivity via two different pathways: a primary pathway where the radioactive debris is taken up directly from the atmosphere, or from precipitation, and a secondary pathway where the radioactivity already deposited in the soil, the sea, fresh water, or in vegetable and animal tissues (from foregoing steps of the food chain) is assimilated. We thus assume that the concentration of a radionuclide in an environmental sample may be related to the deposition rate (fallout rate) of the nuclide in question and to the accumulated amount of activity previously deposited. This basic model was suggested already in the first UNSCEAR report in 1958 (Un58); later, the models have been further developed. The aim of the prediction models in this study was primarily to estimate the transfer factor P_{23} (Un58-77) from fallout in the various samples studied, i.e. the radioecological sensitivity. Is was thus not a main purpose to explain the numerous processes acting in the transfer of the radioactive contamination although such processes have been dealt with throughout the text. Hence the expression for the ^{90}Sr content of inner Danish surface waters (cf.table C.1.5.1)does not, e.g., give any information on the inflow or outflow from other compartments; it merely relates the concentration in the water to the deposition data over Denmark, although a substantial part of the ^{90}Sr in the water has its origin outside Denmark. Dynamic modelling techniques, or systems

simulations (Eb67), are especially useful - even though they also contain "black boxes" - when the responses to variations in the various compartments are studied. However, in the case of global fallout where the source of contamination is common, models such as those applied in this study have generally been preferred (Un58-77).

C.2. Actual models

At present UNSCEAR (Un77) uses a model of the form:

$$Y_i = a \cdot d_i + b \cdot d_{i-1} + c \cdot A_{i-2\ (n)} \qquad \text{(Eq.C.1)}$$

where Y_i is the concentration of a given radionuclide in a given sample collected in the year (i), d_i and d_{i-1} are the fallout rates (in mCi km^{-2}) in the years (i) and (i-1), respectively, of the radionuclide in question, and $A_{i-2\ (n)}$ is the accumulated fallout by the year (i-2) assuming an effective halflife of n years, which at most equals the radiological halflife of the radionuclide, but is usually lower; a is the so-called rate factor, b the lag rate factor and c the soil factor.

The model applied in this study is a further development of the UNSCEAR model:

$$Y_i = a \cdot d_i + b \cdot d_{i-1} + c \cdot A_{i-2\ (n)} + d \cdot A_{i-2\ (R)}, \qquad \text{(Eq.C.2)}$$

the last term in this expression makes allowance for the fact that the effective halflife of the radionuclide probably changes in time and may ultimately approach the radiological halflife (R) (cf.1.6); d is the ultimate soil factor.

The two models mentioned above have been used especially for ^{90}Sr, but in some samples they have also been applied to ^{137}Cs. However, as the root uptake of ^{137}Cs from Danish soils is often negligible, a third model was applied especially for this

radionuclide:

$$Y'_i = a \cdot d'_i + b \cdot d'_{i-1} + f \cdot d'_{i-2}. \qquad \text{(Eq.C.3)}$$

To distinguish the d_i and A_i values for ^{90}Sr and ^{137}Cs, respectively, the ^{137}Cs values are marked by an apostrophe: d'_i, A'_i, etc; no apostrophe always indicates ^{90}Sr data, and two apostrophes are used for ^{54}Mn (cf.table C.2.2.7).

Modified models were applied for ^{90}Sr and ^{137}Cs in grain:

$$^{90}Sr : Y_i = a_1 \cdot d_{i\ (J-A)} + c\ A_{i-1\ (n)} \qquad \text{(Eq.C.4)}$$

$$Y_i = a_1 \cdot d_{i\ (J-A)} + c\ A_{i-1\ (n)} + d\ A_{i-1\ (R)} \qquad \text{(Eq.C.5)}$$

and ^{137}Cs: $Y'_i = a_2 \cdot d'_{i\ (M-A)}$ (Eq.C.6)

where $d_{i\ (J-A)}$ is mCi ^{90}Sr km^{-2} deposited in July-August of the year (i) and $d'_{i\ (M-A)}$ is mCi ^{137}Cs km^{-2} deposited in May-August of the year (i). a_1 and a_2 are the corresponding rate factors.

The factors (a, b, c, ...) in the equations have been calculated by a least squares procedure. The squared sum S_t of the differences between calculated and observed values (y_i) over the period of observations (p years) was minimized, e.g.:

$$S_t = \sum_{i=1}^{i=p} (a\ d_i + b \cdot d_{i-1} + c \cdot A_{i-2\ (n)} + d \cdot A_{i-2\ (R)} - y_i)^2.$$

If $d_i = x_1$, $d_{i-1} = x_2$, $A_{i-2\ (n)} = x_3$, $A_{i-2\ (R)} = x_4$ and $y_i = y$, then

$$\frac{\delta S}{\delta a} = a\Sigma x_1^2 + b\Sigma x_1 x_2 + c\Sigma x_1 x_3 + d\Sigma x_1 x_4 - \Sigma x_1 y = 0$$

$$\frac{\delta S}{\delta b} = b\Sigma x_2^2 + a\Sigma x_1 x_2 + c\Sigma x_2 x_3 + d\Sigma x_2 x_4 - \Sigma x_2 y = 0$$

$$\frac{\delta S}{\delta c} = c\Sigma x_3^2 + a\Sigma x_1 x_3 + b\Sigma x_2 x_3 + d\Sigma x_3 x_4 - \Sigma x_3 y = 0$$

$$\frac{\delta S}{\delta d} = d\Sigma x_4^2 + a\Sigma x_1 x_4 + b\Sigma x_2 x_4 + c\Sigma x_3 x_4 - \Sigma x_4 y = 0$$

These four equations may be solved for given effective (n) and radiological (R) halflives. A computer program (Li75) FIT was developed which, for a fixed R, e.g. for ^{90}Sr (R = 28 years), tests all effective halflives (n) from 1-27 years. The positive coefficients of a, b, c ... and the value of n which give the highest correlation coefficient (r) between observed and calculated values are considered as the best estimate. In the evaluation of the quality of the various models, the mean ratio between observed and calculated values is also applied. Thus, if two models have similar correlation coefficients, the model with the mean ratio closest to 1.00 is generally preferred.

C.3. Radioecological sensitivity

The prediction models serve a dual purpose. As their name implies, they are used for predictions of the concentrations of radionuclides in various samples from fallout data; but they may also, as mentioned above, be applied to estimate the transfer coefficient from one step in the food chain to the next. As a special case of the latter purpose, the models may be used for the estimation of the infinite time exposure integral from a deposition of 1 mCi km^{-2}, distributed like global fallout throughout a year, to a given item of the food chain.

This quantity is called the radioecological sensitivity, and it is equal to the transfer coefficient from fallout to the sample in question. The transfer coefficient is defined as the ratio of the infinite integral of concentrations in the sample to the ingegrated deposition rate. As $\sum_{m=1}^{\infty} e^{-m\mu} = \frac{e^{-\mu}}{1-e^{-\mu}}$ the infinite time integrals of the models (Eq.C.1-C.6) were:

(1) : $a + b + c \cdot \frac{e^{-\mu}}{1-e^{-\mu}}$

$$(2) : a + b + c \cdot \frac{e^{-\mu}}{1-e^{-\mu}} + d \cdot \frac{e^{-\lambda}}{1-e^{-\lambda}}$$

$$(3) : a + b + f$$

$$(4) : a_1 \cdot 0.24 + c \frac{e^{-\mu}}{1-e^{-\mu}}$$

$$(5) : a_1 \cdot 0.24 + c \frac{e^{-\mu}}{1-e^{-\mu}} + d \frac{e^{-\lambda}}{1-e^{-\lambda}}$$

$$(6) : a_2 \cdot 0.54$$

Where $\mu = \frac{n}{\ln 2}$ and $\lambda = \frac{R}{\ln 2}$

An annual deposit of 1 mCi km^{-2} implies a deposit of 0.24 mCi km^{-2} in July-August and of 0.54 mCi km^{-2} in May-August. These fractions were the average values for the fallout observations made in Denmark since 1962.

The radioecological sensitivity depends upon three variables: radionuclide, sample type and sample location. A comparison with respect to one of these variables implies that the other two are kept constant. Thus a comparison of, e.g., local radioecological sensitivities implies one radionuclide in identical sample types from various locations. It is assumed that the radioecological sensitivity is time independent, although this may be a simplification (Aa72a).

C.4. FIT tables

In the following the prediction models estimated for the various types of sample have been compiled. In general, two models will be given for each sample and each nuclide. For ^{90}Sr, Eqs.C.1 and C.2 (for grain products Eqs.C.4 and C.5) are shown. For ^{137}Cs, Eqs.C.1, C.2 or C.3 (for grain products C.6) are given. For certain samples (e.g. human bone), the estimate of the radioecological sensitivity depended on the model used. In general, Eq.C.2 yielded higher values than Eqs.C.1 and C.3, because the long halflives of ^{90}Sr and ^{137}Cs made the contribution from the last term in Eq.C.2 relatively large.

The tables are numbered as the anova tables in the preceding appendix, which means they show a C followed by three digits representing the chapter and section of the main text and a serial number. In the table each equation has an equation number. For practical reasons, some tables have been subdivided into an a, b (and c) part. When referring to such tables, the a, b (and c) are omitted. Besides the prediction equations, the tables indicate the radioecological sensitivity and the correlation coefficient (r) (Eq.B.7) between the observed data and the values calculated from the equation. The significance level of the correlation coefficient is as usual indicated by stars.

Note added in proof

Since the prediction models were calculated, data from 1977-1979 have become available. These data may be used for a test of the reliability of the predictions from the models.

The relative standard deviation of the ratios between predicted and observed annual values during 1977-1979 was 35%, on the average. Ratios based on models with much data, such as Danish dried milk, showed a relative standard deviation of half of this average. In most cases, the models with three terms (Eq C.1.) gave just as good predictions as those with four terms (Eq C.2.).

The ^{137}Cs models generally underestimated the levels in 1977-1979, probably because the ^{137}Cs deposition was calculated from the ^{90}Sr fallout by multiplication with 1.6. This may have been too low a $^{137}Cs/^{90}Sr$ ratio in 1977-1979, as suggested by the higher ratios found in air samples from these years.

In the case of ^{90}Sr, the ratios between predicted and observed values showed no systematic deviation from unity on the whole. The models for vegetables, milk, meat, bread and total diet tended however to predict higher ^{90}Sr values than observed, whereas the opposite was the case for the models for grain, apples, and eggs.

Table C.1.4.1. Prediction models for ^{90}Sr in fresh water samples (unit for radioecological sensitivity: pCi ^{90}Sr $l^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $l^{-1}_{(i)}$	Sensitivity	r
1	Ground water	Denmark	1961-76	$10^{-3}[1.10d_i+0.91d_{i-1}+0.30A_{i-2(11)}+0.016A_{i-2(28)}]$	7.4×10^{-3}	0.8248***
2	- " -	- " -	- " -	$10^{-3}[1.10d_i+0.91d_{i-1}+0.32A_{i-2(11)}]$	6.9×10^{-3}	0.8248***
3	Stream water	- " -	1964-75[o]	$0.17d_i+0.0028A_{i-1(6)}+0.0035A_{i-1(28)}$	0.33	0.9884***
4	- " -	- " -	- " -	$0.17d_i+0.0059A_{i-1(14)}$	0.29	0.9883***
5	Lake water	- " -	1963-75[p]	$0.18d_i+0.046d_{i-1}+0.22A_{i-2(2)}$	0.76	0.9815**
6	- " -	- " -	- " -	$0.25d_i+0.022A_{i-1(28)}$	1.12	0.9994***
7	Drinking water	- " -	1965-73[q]	$10^{-3}[0.41d_i+0.41A_{i-1(7)}]$	4.4×10^{-3}	0.6515*
8	- " -	- " -	- " -	$10^{-3}[0.74A_{i(3)}+0.061A_{i(28)}]$	5.3×10^{-3}	0.6518*
9	Drinking water	Thorshavn	1962-76	$0.049d_{iF}+0.011d_{i-1F}+0.044A_{i-2(1)F}+0.004A_{i-2(28)F}$	0.36	0.8764***
10	- " -	Klaksvig	- " -	$0.031d_{iF}+0.003A_{i-1(6)F}$	0.096	0.7684***
11	- " -	Tvarå	- " -	$0.025d_{iF}+0.0048A_{i-1(5)F}+0.0004A_{i-1(28)F}$	0.073	0.7370***
12	- " -	W.Greenland	- " -	$0.056d_{iG}+0.048d_{i-1G}+0.020A_{i-2(1)G}+0.010A_{i-2(28)G}$	0.52	0.7963***
13	- " -	- " -	- " -	$0.054d_{iG}+0.058d_{i-1G}+0.011A_{i-2(26)G}$	0.52	0.7853***

For a mean deposition of 1 mCi ^{90}Sr km^{-2} in the Faroes, the depositions in Thorshavn, Klaksvig and Tvarå were 0.67, 1.33 and 0.83 mCi ^{90}Sr km^{-2}, respectively. The depositions at Thorshavn and Klaksvig were calculated from the precipitation data collected in 1962-1976 (RRF 62-76) and the mean deposition in the Faroes was estimated as the mean of these two locations. The deposition at Tvarå was estimated from the ^{90}Sr determination of a soil sample collected in 1967 (cf. Table D.1.6.1.2); the radioecological sensitivity of Faroese drinking water was estimated at: $\frac{0.67\times0.26 + 1.33\times0.056 + 0.83\times0.07}{2.83} = 0.11$.

o) Except 1965-1966, 1969, 1972, 1974.
p) Except 1964-1970, 1972, 1974.
q) Except 1971.

Table C.1.5.1. Prediction models for ^{90}Sr in sea water (unit for radioecological sensitivity: pCi ^{90}Sr $l^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $l^{-1}_{(i)}$	Sensitivity	r
1	Surface water	Inner Danish waters	1961-72[a]	$0.026d_i+0.024d_{i-1}+0.013A_{i-2(1)}+0.0098A_{i-2(28)}$	0.45	0.9624***
2	- " -	- " -	- " -	$0.024d_i+0.027d_{i-1}+0.014A_{i-2(10)}$	0.28	0.9584***
3	- " -	N.Atlantic[b]	1962-76	$10^{-3}[6.9d_{iF}+4.9d_{i-1F}+0.53A_{i-2(10)F}+0.34A_{i-2(28)F}]$	0.033	0.9313***
4	- " -	- " -	- " -	$10^{-3}[6.9d_{iF}+5.0d_{i-1F}+0.85A_{i-2(15)F}]$	0.030	0.9314***
5	- " -	W.Greenland	1962-75	$0.060d_{iG}+0.008d_{i-1G}+0.026A_{i-2(1)G}+0.0033A_{i-2(28)G}$	0.23	0.9605***
6	- " -	E.Greenland	- " -	$0.061d_{iG}+0.006d_{i-1G}+0.008A_{i-2(28)G}$	0.39	0.9176***

a) Due to the inflow of North Sea water contaminated with non-fallout ^{90}Sr after 1972, the succeeding years were omitted.

b) Including Faroese waters. The ^{90}Sr fallout data were the Faroese data.

The fallout data applied to W.Greenland sea water were those from Godthåb; for E.Greenland, data from Kap Tobin were used.

Ocean water (salinity 35‰) contains 0.41 g Ca l^{-1} (Go 71). The calcium content for other salinities (S) may be obtained as: $Ca_S = S \times \frac{0.41}{35}$ g l^{-1}; the calcium contents were used for the estimation of pCi ^{90}Sr (g Ca)^{-1}y per mCi ^{90}Sr km^{-2}, a quantity useful in food chain comparisons.

Table C.2.2.1.a. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye	Jutland	1959-76	$90d_{i(Jul-Aug)}+2.5A_{i-1(1)}+0.21A_{i-1(28)}$	33	0.9926***
2	- " -	- " -	- " -	$90d_{i(Jul-Aug)}+4.1A_{i-1(1)}$	26	0.9910***
4	Barley	- " -	- " -	$81d_{i(Jul-Aug)}+1.34A_{i-1(1)}+0.32A_{i-1(28)}$	34	0.9945***
5	- " -	- " -	- " -	$82d_{i(Jul-Aug)}+0.56A_{i-1(10)}$	28	0.9940***
8	Wheat	- " -	- " -	$68d_{i(Jul-Aug)}+0.67A_{i-1(10)}+0.0076A_{i-1(28)}$	26	0.9916***
9	- " -	- " -	- " -	$68d_{i(Jul-Aug)}+0.68A_{i-1(10)}$	26	0.9916***
12	Oats	- " -	- " -	$57d_{i(Jul-Aug)}+0.74A_{i-1(28)}$	43	0.9824***

Table C.2.2.1.b. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
3	Rye	Islands	1959-76	$89d_{i(Jul-Aug)}+0.067A_{i-1(28)}$	24	0.9797***
6	Barley	- " -	- " -	$53d_{i(Jul-Aug)}+0.25A_{i-1(13)}+0.026A_{i-1(28)}$	18	0.9909***
7	- " -	- " -	- " -	$53d_{i(Jul-Aug)}+0.27A_{i-1(14)}$	18	0.9909***
10	Wheat	- " -	- " -	$57d_{i(Jul-Aug)}+0.43A_{i-1(7)}+0.046A_{i-1(28)}$	20	0.9875***
11	- " -	- " -	- " -	$57d_{i(Jul-Aug)}+0.47A_{i-1(8)}$	19	0.9876***
13	Oats	- " -	- " -	$53d_{i(Jul-Aug)}+0.43A_{i-1(28)}$	30	0.9807***

The radioecological sensitivity of grain in Jutland was 1.44±0.18 (1SD) times that in the Islands.

Table C.2.2.2.a. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Rye	Jutland	1959-76	$221d_{i\,(Jul-Aug)} + 1.68A_{i-1\,(1)} + 0.78A_{i-1\,(28)}$	87	0.9946***
2	- " -	- " -	- " -	$223d_{i\,(Jul-Aug)} + 0.87A_{i-1\,(28)}$	88	0.9945***
4	Barley	- " -	- " -	$162d_{i\,(Jul-Aug)} + 4.9A_{i-1\,(1)} + 0.68A_{i-1\,(28)}$	71	0.9938***
5	- " -	- " -	- " -	$164d_{i\,(Jul-Aug)} + 1.87A_{i-1\,(6)}$	58	0.9927***
8	Wheat	- " -	- " -	$164d_{i\,(Jul-Aug)} + 1.67A_{i-1\,(3)} + 0.89A_{i-1\,(28)}$	82	0.9889***
9	- " -	- " -	- " -	$164d_{i\,(Jul-Aug)} + 2.14A_{i-1\,(8)}$	63	0.9888***
12	Oats	- " -	- " -	$74d_{i\,(Jul-Aug)} + 0.95A_{i-1\,(21)} + 0.003A_{i-1\,(28)}$	46	0.9797***
13	- " -	- " -	- " -	$74d_{i\,(Jul-Aug)} + 0.95A_{i-1\,(21)}$	46	0.9797***

Table C.2.2.2.b. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
3	Rye	Islands	1959-76	$170d_{i\,(Jul-Aug)} + 0.59A_{i-1\,(28)}$	64	0.9907***
6	Barley	- " -	- " -	$98d_{i\,(Jul-Aug)} + 0.90A_{i-1\,(6)} + 0.082A_{i-1\,(28)}$	34	0.9917***
7	- " -	- " -	- " -	$98d_{i\,(Jul-Aug)} + 1.05A_{i-1\,(6)}$	32	0.9917***
10	Wheat	- " -	- " -	$138d_{i\,(Jul-Aug)} + 0.94A_{i-1\,(11)} + 0.048A_{i-1\,(28)}$	50	0.9871***
11	- " -	- " -	- " -	$138d_{i\,(Jul-Aug)} + 1.00A_{i-1\,(11)}$	49	0.9871***
14	Oats	- " -	- " -	$59d_{i\,(Jul-Aug)} + 0.46A_{i-1\,(23)} + 0.058A_{i-1\,(28)}$	32	0.9757***
15	- " -	- " -	- " -	$59d_{i\,(Jul-Aug)} + 0.51A_{i-1\,(25)}$	33	0.9758***

Table C.2.2.1.1.a. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye	Denmark	1959-76	$92d_{i\,(July-Aug)} + 0.072A_{i-1\,(1)} + 0.24A_{i-1\,(28)}$	32	0.9953***
2	- " -	- " -	- " -	$92d_{i\,(July-Aug)} + 0.24A_{i-1\,(28)}$	32	0.9952***
3	Barley	- " -	- " -	$68d_{i\,(July-Aug)} + 0.52A_{i-1\,(1)} + 0.29A_{i-1\,(28)}$	29	0.9965***
4	- " -	- " -	- " -	$68d_{i\,(July-Aug)} + 0.40A_{i-1\,(13)}$	24	0.9964***
5	Wheat	- " -	- " -	$61d_{i\,(July-Aug)} + 0.45A_{i-1\,(11)} + 0.051A_{i-1\,(28)}$	24	0.9967***
6	- " -	- " -	- " -	$61d_{i\,(July-Aug)} + 0.58A_{i-1\,(12)}$	23	0.9967***
7	Oats	- " -	- " -	$56d_{i\,(July-Aug)} + 0.59A_{i-1\,(28)}$	37	0.9949***

Table C.2.2.1.1.b. Prediction models for ^{90}Sr in cereal grain (unit for radioecological sensitivity: pCi ^{90}Sr $(g\ Ca)^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $(g\ Ca)^{-1}_{(i)}$	Sensitivity	r
8	Rye	Denmark	1959-76	$202d_{i\,(July-Aug)} + 0.135A_{i-1\,(1)} + 0.77A_{i-1\,(28)}$	79	0.9958***
9	- " -	- " -	- " -	$203d_{i\,(July-Aug)} + 0.77A_{i-1\,(28)}$	79	0.9958***
10	Barley	- " -	- " -	$132d_{i\,(July-Aug)} + 2.9A_{i-1\,(1)} + 0.61A_{i-1\,(28)}$	59	0.9964***
11	- " -	- " -	- " -	$133d_{i\,(July-Aug)} + 1.36A_{i-1\,(7)}$	45	0.9960***
12	Wheat	- " -	- " -	$147d_{i\,(July-Aug)} + 1.31A_{i-1\,(12)} + 0.018A_{i-1\,(28)}$	59	0.9946***
13	- " -	- " -	- " -	$147d_{i\,(July-Aug)} + 1.34A_{i-1\,(12)}$	58	0.9946***
14	Oats	- " -	- " -	$67d_{i\,(July-Aug)} + 0.69A_{i-1\,(23)} + 0.046A_{i-1\,(28)}$	41	0.9928***
15	- " -	- " -	- " -	$67d_{i\,(July-Aug)} + 0.73A_{i-1\,(24)}$	41	0.9928***

Table C.2.2.4. Prediction models for ^{137}Cs in cereal grain (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye	Jutland	1962-76	$74d^{i}_{i\,(May-Aug)} + 9.2d^{i}_{i-1}$	49	0,9983***
2	- " -	- " -	- " -	$86d^{i}_{i\,(May-Aug)}$	47	0,9879***
3	- " -	Islands	- " -	$78d^{i}_{i\,(May-Aug)}$	42	0,9840***
4	Barley	Jutland	- " -	$63d^{i}_{i\,(May-Aug)}$	34	0,9930***
5	- " -	Islands	- " -	$52.5d^{i}_{i\,(May-Aug)}$	28	0,9931***
6	Wheat	Jutland	- " -	$63.5d^{i}_{i\,(May-Aug)}$	34	0,9830***
7	- " -	Islands	- " -	$45d^{i}_{i\,(May-Aug)}$	24	0,9945***
8	Oats	Jutland	- " -	$52d^{i}_{i\,(May-Aug)}$	28	0,9953***
9	- " -	Islands	- " -	$49d^{i}_{i\,(May-Aug)}$	26	0,9932***

The radioecological sensitivity of grain in Jutland was 1.21±0.15 (1SD) times that in the Islands.

Table C.2.2.5. Prediction models for ^{137}Cs in cereal grain (unit for radioecological sensitivity: pCi ^{137}Cs $(g\ K)^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $(g\ K)^{-1}_{(i)}$	Sensitivity	r
1	Rye	Jutland	1962-76	$20.5d^{i}_{i\,(May-Aug)} + 0.95d^{i}_{i-1}$	12.0	0,9979***
2	- " -	- " -	- " -	$22d^{i}_{i\,(May-Aug)}$	11.8	0,9959***
3	- " -	Islands	- " -	$17.2d^{i}_{i\,(May-Aug)}$	9.3	0,9967***
4	Barley	Jutland	- " -	$15.4d^{i}_{i\,(May-Aug)}$	8.3	0,9898***
5	- " -	Islands	- " -	$12.0d^{i}_{i\,(May-Aug)}$	6.5	0,9862***
6	Wheat	Jutland	- " -	$18.5d^{i}_{i\,(May-Aug)}$	10.0	0,9722***
7	- " -	Islands	- " -	$12.9d^{i}_{i\,(May-Aug)}$	7.0	0,9894***
8	Oats	Jutland	- " -	$14.0d^{i}_{i\,(May-Aug)}$	7.6	0,9841***
9	- " -	Islands	- " -	$13.0d^{i}_{i\,(May-Aug)}$	7.0	0,9888***

Table C.2.2.6.a. Prediction models for ^{137}Cs in cereal grain (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye	Denmark	1962-76	$79d^{\cdot}_{i\,(May-Aug)}+5.0d^{\cdot}_{i-1}$	47	0.9991***
2	- " -	- " -	- " -	$85d^{\cdot}_{i\,(May-Aug)}$	46	0.9957***
3	Barley	- " -	- " -	$59d^{\cdot}_{i\,(May-Aug)}$	32	0.9957***
4	Wheat	- " -	- " -	$52d^{\cdot}_{i\,(May-Aug)}$	28	0.9857***
5	Oats	- " -	- " -	$50d^{\cdot}_{i\,(May-Aug)}$	27	0.9952***

Table C.2.2.6.b. Prediction models for ^{137}Cs in cereal grain (unit for radioecological sensitivity: pCi ^{137}Cs $(g\ K)^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $(g\ K)^{-1}_{(i)}$	Sensitivity	r
6	Rye	Denmark	1962-76	$19.5d^{\cdot}_{i\,(May-Aug)}+0.88d^{\cdot}_{i-1}$	11.4	0.9991***
7	- " -	- " -	- " -	$20.6d^{\cdot}_{i\,(May-Aug)}$	11.1	0.9972***
8	Barley	- " -	- " -	$14.1d^{\cdot}_{i\,(May-Aug)}$	7.6	0.9903***
9	Wheat	- " -	- " -	$15.0d^{\cdot}_{i\,(May-Aug)}$	8.1	0.9777***
10	Oats	- " -	- " -	$13.6d^{\cdot}_{i\,(May-Aug)}$	7.3	0.9884***

Table C.2.2.7. Prediction models for ^{54}Mn in cereal grain (unit for radioecological sensitivity: pCi ^{54}Mn $kg^{-1}y$ per mCi ^{54}Mn km^{-2} a))

No.	Sample	Area	Period	^{54}Mn-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye	Denmark	1963-65	$62d^{"}_{i\,(May-Aug)}$	33	0.9957
2	- " -	- " -	- " -	$27d^{"}_{(Sep(i-1)-Aug(i))}$	27	0.9973°
3	Barley	- " -	- " -	$54d^{"}_{i\,(May-Aug)}$	29	0.9936
4	- " -	- " -	- " -	$23d^{"}_{(Sep(i-1)-Aug(i))}$	23	0.9987°
5	Wheat	- " -	- " -	$41d^{"}_{i\,(May-Aug)}$	32	0.9994°
6	- " -	- " -	- " -	$18d^{"}_{(Sep(i-1)-Aug(i))}$	18	0.9916
7	Oats	- " -	- " -	$54d^{"}_{i\,(May-Aug)}$	29	0.9973°
8	- " -	- " -	- " -	$23d^{"}_{(Sep(i-1)-Aug(i))}$	23	0.9958

a) The ^{54}Mn fallout rate was estimated from the ^{54}Mn air levels measured at Risø, assuming a washout factor of 0.99 pCi l^{-1} per fCi m^{-3} (1.3.3), and using the mean amounts of precipitation observed at the state experimental farms where the crops were collected.

Table C.2.3.1.a. Prediction models for ^{90}Sr in bread and grits (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Rye bread, dark	Denmark[o]	1962-76	$63d_{i-1(Jul-Aug)}+0.19A_{i-2(28)}$	23	0.9899***
2	White bread	- " -	- " -	$7.1d_{i-1(Jul-Aug)}+0.23d_{i-2}+0.36A_{i-3(1)}+0.056A_{i-3(28)}$	5.0	0.9878***
3	- " -	- " -	- " -	$7.0d_{i-1(Jul-Aug)}+0.29d_{i-2}+0.19A_{i-3(5)}$	3.2	0.9865***
6	Rye bread, light	Faroes[p]	1964-75	$6.4d_{i-1}+0.22d_{i-2}+0.18A_{i-3(28)}$	14[q]	0.9887***
7	White bread	- " -	- " -	$0.66d_{i-1}+0.25d_{i-2}+0.068A_{i-3(28)}$	3.6[q]	0.9023***
10	Oat grits	Denmark	1960-72[x]	$11.6d_{i-1(Jul-Aug)}+0.29A_{i-2(28)}$	14.3	0.9517***

o) The Danish bread samples were collected in the 8 zones in June.

p) The Faroese bread was sampled in Thorshavn in June and December.

q) The sensitivities calculated for Faroese bread were based on Danish fallout data, because the flour was imported.

x) Except 1961, 1962 and 1967-1969.

Table C.2.3.1.b. Prediction models for ^{137}Cs in bread and grits (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
4	Rye bread, dark	Denmark[o]	1964-75	$31d'_{i-1}+3.3d'_{i-2}+3.4d'_{i-3}$	38	0.9985***
5	White bread	- " -	- " -	$7.1d'_{i-1}+3.1d'_{i-2}$	10	0.9913***
8	Rye bread, light	Faroes[p]	1964-75	$23d'_{i-1}+0.58d'_{i-2}+0.43d'_{i-3}$	24[q]	0.9922***
9	White bread	- " -	- " -	$5.5d'_{i-1}+3.0d'_{i-2}$	8.5[q]	0.9639***
11	Oat grits	Denmark	1963-72[x]	$24d'_{i-1(May-Aug)}$	13	0.8968**

o) The Danish bread samples were collected in the 8 zones in June.

p) The Faroese bread was sampled in Thorshavn in June and December.

q) The sensitivities calculated for Faroese bread were based on Danish fallout data, because the flour was imported.

x) Except 1961, 1962 and 1967-1969.

Table C.2.4.1. Prediction models for ^{90}Sr in grass (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sampling season	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Whole year	Zealand	1960-76	$29d_i+13d_{i-1}+4.3A_{i-2(1)}+0.21A_{i-2(28)}$	55	0.9910***
2	- " -	- " -	- " -	$29d_i+12.5d_{i-1}+5.9A_{i-2(1)}$	47	0.9910***
3	Summer half-year	Denmark	1962-70	$28d_i+2.9A_{i-1(1)}$	31	0.9316***
4	- " -	Faroes	1962-75	$169d_{iF}+0.12d_{i-1F}+10.7A_{i-2(1)F}+1.36A_{i-2(28)F}$	234	0.9734***
5	- " -	- " -	- " -	$167d_{iF}+1.25d_{i-1F}+5.7A_{i-2(4)F}$	198	0.9745***
6	- " -	West Greenland	1962-73[a)]	$111d_{iG}+9.9d_{i-1G}+15A_{i-2(14)G}+0.78A_{i-2(28)G}$	448	0.5969*
7	- " -	- " -	- " -	$111d_{iG}+9.7d_{i-1G}+16A_{i-2(14)G}$	436	0.5970*

a) Except 1971.

The radioecological sensitivity per kg^{-1} fresh weight was estimated from the Danish Ca concentration in fresh grass, i.e. 1.24 g Ca kg^{-1}, assuming fresh grass in the Faroes and Greenland to have the same Ca-concentration.

Table C.2.4.2.a. Prediction models for ^{137}Cs in summer grass (unit for radioecological sensitivity: pCi ^{137}Cs $(g\ K)^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sampling season	Area	Period	^{137}Cs-activity, pCi $(g\ K)^{-1}_{(i)}$	Sensitivity	r
1	Summer half-year	Denmark	1964-70	$3.25d'_i+0.033d'_{i-1}+0.042A'_{i-2(30)}$	5.1	0.9693***
2	- " -	- " -	- " -	$3.24d'_i+0.043A'_{i-1(30)}$	5.1	0.9693***
3	- " -	Faroes	1964-75	$64A'_{i(1)F}$	64	0.8279***
4	- " -	- " -	- " -	$87d'_{iF}$	87	0.9510***
5	- " -	W.Greenland	1964-73 o)	$75A'_{i(1)G}$	75	0.4547
6	- " -	- " -	- " -	$82d'_{iG}$	82	0.3329

o) Except 1971.

Table C.2.4.2.b. Prediction models for ^{137}Cs in summer grass (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sampling season	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
7	Summer half-year	Denmark	1964-70	$16d'_i+0.70A'_{i-1(2)}+0.23A'_{i-1(30)}$	27	0.9906***
8	- " -	- " -	- " -	$16d'_i+0.26d'_{i-1}+0.62A'_{i-2(5)}$	20	0.9905***

Danish grass was available as fresh samples, and prediction models were therefore also calculated per kg^{-1} fresh weight.
The radioecological sensitivity per kg^{-1} fresh weight for Faroese and Greenlandic grass was estimated from the K-concentrations in grass (D.2.4.1), and under the assumption that fresh grass in Greenland and the Faroes contained 1.24 g Ca kg^{-1} fresh weight, i.e. the same concentration as Danish grass.

Table C.2.4.3. Prediction models for ^{90}Sr in various fodder crops (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Roots	Denmark	1959-74	$1.53d_i+0.45d_{i-1}+0.62A_{i-2(11)}+0.059A_{i-2(28)}$	13.9	0.7012**
2	- " -	- " -	- " -	$1.53d_i+0.45d_{i-1}+0.67A_{i-2(12)}$	13.2	0.7013**
3	Leaves	- " -	1962-74	$13.5d_i+0.43d_{i-1}+2.4A_{i-2(28)}$	111	0.8757***
4	Straw	- " -	1959-76	$680d_{i(Jul-Aug)}+5.2A_{i-1(1)}+2.9A_{i-1(28)}$	284	-
5	- " -	- " -	- " -	$683d_{i(Jul-Aug)}+4.0A_{i-1(13)}$	237	-

Roots and leaves were based on samples of swedes and beets. The missing values were estimated by VAR 3 by means of data on potatoes and carrots for the roots and of data on kale and cabbage for the leaves (RRD 74).
In the calculations it was assumed that roots on the average contained 0.4 g Ca, 2 g K and 0.11 kg dry matter per kg fresh weight (Br 69), (An 67b), and that leaves contained 2.4 g Ca, 3 g K and approx. 0.1 kg dry matter per kg fresh weight (Br 69), (An 67a).
Experimentally it has been shown that the concentration of radiostrontium in straw from direct contamination (VI), (VIII) as well as from indirect contamination (RRD 59-76) is an order of magnitude higher than in grain. Hence the prediction models for straw were identical to those for barley grain multiplied by a factor of ten. Straw contained 2.4 g Ca kg^{-1} fresh weight (Br 69).

Table C.2.4.4. Prediction models for ^{137}Cs in various fodder crops (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Roots	Denmark	1963-74	$0.78d'_i$	0.8	0.9780***
2	Leaves	- " -	- " -	$9.7d'_i+0.08A'_{i-1(30)}$	13	0.9681***
3	- " -	- " -	- " -	$10.0d'_i$	10	0.9695***
4	Straw	- " -	- " -	$117d'_{i(May-Aug)}$	63	-

Cf. remarks to Table C.2.4.3.
It has been found experimentally that the radiocesium concentration in straw is twice that in grain (VI), (VIII), (Aa72b). The prediction model for ^{137}Cs in straw was thus estimated as twice the model for barley grain.

Table C.2.5.1.a. Prediction models for ^{90}Sr in vegetables and fruits (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Cabbage	Jutland	1961-75	$0.27d_i+0.66d_{i-1}+0.114A_{i-2(1)}+0.17A_{i-2(28)}$	7.8	0.8865***
2	- " -	- " -	- " -	$0.24d_i+0.72d_{i-1}+0.18A_{i-2(28)}$	8.1	0.8819***
3	- " -	Islands	- " -	$0.56d_i+0.01d_{i-1}+0.08A_{i-2(3)}+0.18A_{i-2(28)}$	8.0	0.8165***
4	- " -	- " -	- " -	$0.56d_i+0.03d_{i-1}+0.23A_{i-2(17)}$	6.1	0.8061***
5	Carrot	Jutland	- " -	$0.16d_i+1.09d_{i-1}+0.29A_{i-2(28)}$	12.8	0.8057***
6	- " -	Islands	- " -	$0.34d_i+0.31A_{i-1(9)}+0.016A_{i-1(28)}$	4.9	0.5282*
7	- " -	- " -	- " -	$0.55d_i+0.001d_{i-1}+0.31A_{i-2(10)}$	4.9	0.5673*

Table C.2.5.1.b. Prediction models for ^{90}Sr in vegetables and fruits (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
8	Potato	Jutland	1959-75	$0.13d_i+0.077d_{i-1}+0.05A_{i-2(1)}+0.054A_{i-2(28)}$	2.4	0.8383***
9	- " -	- " -	- " -	$0.12d_i+0.10d_{i-1}+0.056A_{i-2(28)}$	2.5	0.8301***
10	- " -	Islands	- " -	$0.16d_i+0.073d_{i-1}+0.061A_{i-2(28)}$	2.7	0.9663***
11	- " -	Faroes	1962-76	$0.09d_{iF}+0.59d_{i-1F}+0.27A_{i-2(1)F}+0.058A_{i-2(28)F}$	3.3	0.8596***
12	- " -	- " -	- " -	$0.034d_{iF}+0.71d_{i-1F}+0.097A_{i-2(11)F}$	2.2	0.8388***
13	Apple	Denmark	1959-76	$1.11d_i+0.0072A_{i-1(28)}$	1.4	0.8692***

The fallout-weighted (cf. Table C.3.2.1) mean sensitivities for Denmark became: 7.6 for cabbage, 9.3 for carrots and 2.6 for potatoes. Faroese potatoes showed a mean sensitivity of 2.8.

Table C.2.5.2.a. Prediction models for ^{90}Sr in vegetables (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Cabbage	Jutland	1961-75	$0.63d_i+1.37d_{i-1}+0.39A_{i-2(28)}$	17.6	0.7816***
2	- " -	Islands	- " -	$0.86d_i+0.23d_{i-1}+0.25A_{i-2(1)}+0.36A_{i-2(28)}$	15.8	0.7524***
3	- " -	- " -	- " -	$0.82d_i+0.36d_{i-1}+0.37A_{i-2(28)}$	16.0	0.7426***
4	Carrot	Jutland	- " -	$1.44d_i+1.91A_{i-1(1)}+0.95A_{i-1(28)}$	41	0.7623***
5	- " -	- " -	- " -	$0.60d_i+3.2d_{i-1}+0.95A_{i-1(28)}$	42	0.8001***
6	- " -	Islands	- " -	$1.18d_i+1.18A_{i-1(7)}+0.001A_{i-1(28)}$	12.6	0.5665*
7	- " -	- " -	- " -	$1.85d_i+0.033d_{i-1}+1.13A_{i-2(7)}$	12.7	0.6060*

Table C.2.5.2.b. Prediction models for ^{90}Sr in vegetables and fruits (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
8	Potato	Jutland	1959-75	$2.6d_i+2.3A_{i-1(1)}+1.22A_{i-1(28)}$	54	0.5991**
9	- " -	- " -	- " -	$1.8d_i+3.9d_{i-1}+1.23A_{i-1(28)}$	55	0.6597**
10	- " -	Islands	- " -	$1.6d_i+1.7d_{i-1}+1.15A_{i-2(28)}$	49	0.5500*
11	- " -	Faroes	1962-76	$5.8d_{iF}+17.8d_{i-1F}+2.6A_{i-2(21)F}+0.062A_{i-2(28)F}$	105	0.7667***
12	- " -	- " -	- " -	$5.8d_{iF}+17.7d_{i-1F}+2.7A_{i-2(20)F}$	100	0.7667***
13	Apple	Denmark	1959-76	$21d_i+0.14A_{i-1(28)}$	27	0.9132***

The fallout-weighted (cf. Table C.3.2.1) mean sensitivities for Denmark became: 16.9 for cabbage, 29 for carrots and 52 for potatoes. Faroese potatoes showed a mean sensitivity of 102.

Table C.2.5.3. Prediction models for ^{137}Cs in vegetables and fruits (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Cabbage	Denmark	1963-75	$0.81d'_i+0.044A'_{i-1(7)}+0.0015A'_{i-1(30)}$	1.3	0.9009***
2	- " -	- " -	- " -	$0.81d'_i+0.043A'_{i-1(8)}$	1.3	0.9010***
3	Carrot	- " -	- " -	$1.53d'_i+0.011d'_{i-1}+0.052A'_{i-2(3)}$	1.8	0.9856***
4	- " -	- " -	- " -	$1.49d'_i+0.093A'_{i-1(2)}$	1.7	0.9857***
5	Potato	Jutland	- " -	$3.3d'_i+0.038A'_{i-1(30)}$	4.9	0.9900***
6	- " -	- " -	- " -	$3.5d'_i$	3.5	0.9904***
7	- " -	Islands	- " -	$3.3d'_i$	3.3	0.9585***
8	- " -	Faroes	1963-76	$1.67d'_{iF}+0.58d'_{i-1F}+4.9A'_{i-2(2)F}+1.04A'_{i-2(30)F}$	59	0.5622*
9	- " -	- " -	- " -	$1.28d'_{iF}+0.81d'_{i-1F}+4.9A'_{i-2(4)F}$	28	0.5433*
10	Apple	Denmark	1964-76	$2.0d'_i+1.13d'_{i-1}+0.82A'_{i-2(1)}$	3.9	0.9902***
11	- " -	- " -	- " -	$2.3d'_i+1.8A'_{i-1(1)}$	4.1	0.9902***

The fallout-weighted (cf. Table C.3.2.1) mean sensitivities for Denmark became: 1.3 for cabbage, 1.8 for carrots and 3.8 for potatoes. Faroese potatoes showed a mean sensitivity of 44.

Table C.2.5.4. Prediction models for ^{137}Cs in vegetables and fruits (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
1	Cabbage	Denmark	1963-75	$0.32d'_i+0.031A'_{i-1(4)}+0.0012A'_{i-1(30)}$	0.53	0.8935***
2	- " -	- " -	- " -	$0.32d'_i+0.028A'_{i-1(5)}$	0.51	0.8936***
3	Carrot	- " -	- " -	$0.63d'_i+0.021d'_{i-1}+0.036A'_{i-2(2)}$	0.75	0.9829***
4	- " -	- " -	- " -	$0.61d'_i+0.113A'_{i-1(1)}$	0.72	0.9833***
5	Potato	Jutland	- " -	$0.94d'_i+0.004A'_{i-1(30)}$	1.11	0.9849***
6	- " -	- " -	- " -	$0.96d'_i$	0.96	0.9851***
7	- " -	Islands	- " -	$0.94d'_i$	0.94	0.9578***
8	- " -	Faroes	1963-76	$0.043d'_{iF}+0.18d'_{i-1F}+2.2A'_{i-2(2)F}+0.22A'_{i-2(30)F}$	14.9	0.6254*
9	- " -	- " -	- " -	$0.022d'_{iF}+0.92A'_{i-1(10)F}$	12.8	0.5675*
10	Apple	Denmark	1963-76	$4.7d'_i$	4.7	0.9803***

The fallout-weighted (cf. Table C.3.2.1) mean sensitivities for Denmark became: 0.52 for cabbage, 0.74 for carrots and 0.99 for potatoes. Faroese potatoes showed a mean sensitivity of 13.8.

Table C.2.5.5.a. Prediction models for irregularly collected Danish vegetables and fruits (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Kale	Denmark	1960-76[o)]	$3.2d_i+4.6d_{i-1}+0.82A_{i-2(10)}+0.088A_{i-2(28)}$	23	0.9358***
2	Bean	- " -	1960-72[p)]	$1.25d_i+0.49A_{i-1(28)}$	21	0.7879*
3	Pea	- " -	1964-73[q)]	$0.40d_i+0.12d_{i-1}+0.26A_{i-2(5)}$	2.3	0.7720*
4	Onion	- " -	1960-74[x)]	$0.23d_i+0.45d_{i-1}+0.29A_{i-2(28)}$	12.3	0.5435*
5	Strawberry	- " -	1960-71[s)]	$1.4d_i+0.12d_{i-1}+0.16A_{i-2(1)}+0.22A_{i-2(28)}$	10.6	0.8723***

o) Except 1961, 1969-1975.
p) Except 1961, 1965, 1967, 1969-1971.
q) Except 1968, 1969, 1972.
x) Except 1962, 1963.
s) Except 1961, 1963, 1967-1969.

Table C.2.5.5.b. Prediction models for irregularly collected Danish vegetables and fruits (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
6	Kale	Denmark	1963-76[o)]	$6.5d'_i+1.05d'_{i-1}+0.38d'_{i-2}$	7.9	0.9860***
7	Bean	- " -	1964-72[t)]	$2.2d'_i+0.062A'_{i-1(1)}$	2.3	0.9730*
8	Pea	- " -	1963-74[u)]	$1.48d'_i+0.87d'_{i-1}$	2.4	0.9766***
9	Onion	- " -	1963-74	$0.07d'_i+0.144A'_{i-1(4)}$	0.8	0.6826*
10	Strawberry	- " -	1965-71[v)]	$2.6d'_i$	2.6	0.9862**

o) Except 1961, 1969-1975.
t) Except 1963-1965, 1967, 1969-1971.
u) Except 1967, 1968, 1971, 1972.
v) Except 1967-1969.

Table C.2.6.1. Prediction models for ^{90}Sr and ^{137}Cs in W.Greenland lichen (unit for radioecological sensitivity: nCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2} and nCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	Equation	Sensitivity	r
1	Lichen	W.Greenland	1962-75	nCi $^{90}Sr\ kg^{-1}_{(i)} = 0.10d_{iG} + 0.40A_{i-1(3)G}$	1.6	0.2962
2	- " -	- " -	- " -	nCi $^{137}Cs\ kg^{-1}_{(i)} = 0.72d'_{iG} + 0.89d'_{i-1G} + 0.37A'_{i-2(30)G}$	17.6	0.7732**

Table C.2.7.1. Prediction models for ^{90}Sr in sea plants (unit for radioecological sensitivity: pCi ^{90}Sr $(g\ Ca)^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $(g\ Ca)^{-1}_{(i)}$	Sensitivity	r
1	Zostera marina	Zealand	1959-75	$0.17d_i + 0.92d_{i-1} + 0.34A_{i-2(1)} + 0.045A_{i-2(28)}$	3.2	0.9322***
2	- " -	- " -	- " -	$0.13d_i + 1.03d_{i-1} + 0.11A_{i-2(7)}$	2.2	0.9199***
3	Fucus vesiculosus	- " -	1959-75[a]	$0.013d_i + 0.45d_{i-1} + 1.72A_{i-2(1)} + 0.29A_{i-2(28)}$	13.8	0.9221***
4	- " -	- " -	- " -	$0.035d_i + 0.68A_{i-1(8)}$	7.5	0.8428***
5	Fucus and Laminaria	Faroes	1962-76	$0.048d_{iF} + 0.078d_{i-1F} + 0.37A_{i-2(1)F} + 0.028A_{i-2(28)F}$	1.61	0.7540***
6	- " -	- " -	- " -	$0.097d_{iF} + 0.009d_{i-1F} + 0.60A_{i-2(1)F}$	0.71	0.7255**
7	- " -	W.Greenland	1962-76[b]	$0.73d_{iG} + 0.073d_{i-1G} + 0.12A_{i-2(28)G}$	5.6	0.5538*

a) Except 1966 and 1969.

b) Except 1966-69.

Table C.3.2.1. Prediction models for ^{90}Sr in Danish milk (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Dried milk	Jutland	1962-76	$1.53d_i+0.29d_{i-1}+0.24A_{i-2(2)}+0.056A_{i-2(28)}$	4.64	0.9979***
2	- " -	- " -	- " -	$1.50d_i+0.32d_{i-1}+0.21A_{i-2(5)}$	3.23	0.9974***
3	- " -	Islands	- " -	$1.20d_i+0.25d_{i-1}+0.13A_{i-2(5)}+0.012A_{i-2(28)}$	2.80	0.9977***
4	- " -	- " -	- " -	$1.20d_i+0.26d_{i-1}+0.135A_{i-2(6)}$	2.56	0.9978***
5	Fresh milk	Jutland	- " -	$1.22d_i+0.55d_{i-1}+0.14A_{i-2(5)}+0.019A_{i-2(28)}$	3.48	0.9911***
6	- " -	- " -	- " -	$1.22d_i+0.55d_{i-1}+0.16A_{i-2(6)}$	3.08	0.9911***
7	- " -	Islands	- " -	$1.02d_i+0.28d_{i-1}+0.12A_{i-2(3)}+0.040A_{i-2(28)}$	3.36	0.9954***
8	- " -	- " -	- " -	$1.02d_i+0.29d_{i-1}+0.14A_{i-2(6)}$	2.45	0.9953***

If the mean deposition for Denmark is 1 mCi ^{90}Sr km^{-2}, the deposition in Jutland is 1.113 mCi ^{90}Sr km^{-2} and in the Islands 0.887 mCi ^{90}Sr km^{-2}. The transfer factors for the entire country then become: $\frac{1.113\times(4.64+3.23)\ +\ 0.887\times(2.80+2.56)}{4}$ = 3.4 pCi ^{90}Sr (g Ca)$^{-1}$ per mCi ^{90}Sr km^{-2} for dried milk and 3.1 for fresh milk. The dried milk figures were a 12-month mean (milk year), while the fresh milk value was based on the mean of June and December only.

Table C.3.2.2. Prediction models for ^{137}Cs in Danish milk (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
1	Dried milk	Jutland	1962-76	$3.52d'_i+0.17d'_{i-1}+0.13A'_{i-2(2)}+0.0079A'_{i-2(30)}$	4.41	0.9990***
2	- " -	- " -	- " -	$3.52d'_i+0.18d'_{i-1}+0.11A'_{i-2(3)}$	4.17	0.9990***
3	- " -	Islands	- " -	$2.21d'_i+0.35d'_{i-1}+0.019A'_{i-2(9)}+0.0009A'_{i-2(30)}$	2.84	0.9976***
4	- " -	- " -	- " -	$2.21d'_i+0.35d'_{i-1}+0.018A'_{i-2(11)}$	2.85	0.9976***
5	Fresh milk	Jutland	- " -	$2.61d'_i+0.89d'_{i-1}+0.13A'_{i-2(1)}$	3.63	0.9962***
6	- " -	- " -	- " -	$2.67d'_i+0.77d'_{i-1}+0.20d'_{i-2}$	3.64	0.9967***
7	- " -	Islands	- " -	$1.75d'_i+0.66d'_{i-1}+0.026A'_{i-2(4)}+0.0008A'_{i-2(30)}$	2.59	0.9970***
8	- " -	- " -	- " -	$1.76d'_i+0.66d'_{i-1}+0.023A'_{i-2(5)}$	2.58	0.9970***

The transfer factors for the entire country were calculated as shown in table C.3.2.1. For dried milk the radioecological sensitivity became 3.7 and for fresh milk 3.2.

Table C.3.3.1. Prediction models for ^{90}Sr in Faroese milk (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Fresh milk	Faroes	1962-76	$2.87d_{iF}+1.89d_{i-1F}+0.79A_{i-2(2)F}+0.073A_{i-2(28)F}$	9.6	0.9936***
2	- " -	- " -	- " -	$2.82d_{iF}+1.94d_{i-1F}+0.73A_{i-2(3)F}$	6	0.9932***

The fresh milk was collected every week in Thorshavn, Klaksvig and Tvarå.

Table C.3.3.2.a. Prediction models for ^{137}Cs in Faroese milk (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
1	Fresh milk	Thorshavn	1962-76	$7.79d'_{iF}+9.68d'_{i-1F}+3.42A'_{i-2(2)F}+0.23A'_{i-2(30)F}$	35.6	0.9763***
2	- " -	- " -	- " -	$7.60d'_{iF}+9.93d'_{i-1F}+2.93A'_{i-2(3)F}$	28.8	0.9768***
3	- " -	Klaksvig	- " -	$6.28d'_{iF}+4.94d'_{i-1F}+2.50A'_{i-2(3)F}+0.028A'_{i-2(30)F}$	22.0	0.9953***
4	- " -	- " -	- " -	$6.92d'_{iF}+4.89d'_{i-1F}+2.59A'_{i-2(3)F}$	21.8	0.9953***
5	- " -	Tvarå	- " -	$9.74d'_{iF}+10.3d'_{i-1F}+3.88A'_{i-2(2)F}+0.96A'_{i-2(30)F}$	70.5	0.9830***
6	- " -	- " -	- " -	$9.30d'_{iF}+10.9d'_{i-1F}+3.54A'_{i-2(5)F}$	44.0	0.9826***

If the mean deposition for the Faroes is 1 mCi ^{137}Cs km^{-2}, the depositions in Thorshavn, Klaksvig and Tvarå are 0.67, 1.33 and 0.83 mCi ^{137}Cs km^{-2}, respectively (cf. note to Table C 1.4.1.b). The radioecological sensitivity for pCi ^{137}Cs (g K)$^{-1}$ in Faroese milk becomes: $\frac{0.67\times35.6 + 1.33\times22.0 + 0.83\times70.5}{2.83}$ = 39.4(Nos. 1, 3, 5), or 29.8 (Nos. 2, 4, 6).

Table C.3.3.2.b. Prediction models for ^{137}Cs in Faroese milk (unit for radioecological sensitivity: pCi ^{137}Cs l^{-1}y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi l$^{-1}_{(i)}$	Sensitivity	r
7	Fresh milk	Thorshavn	1964-76	$8.17d'_{iF}+20.0d'_{i-1F}+4.56A'_{i-2(3)F}+0.060A'_{i-2(30)F}$	48.3	0.9952***
8	- " -	- " -	- " -	$8.70d'_{iF}+19.7d'_{i-1F}+4.74A'_{i-2(3)F}$	46.6	0.9951***
9	- " -	Klaksvig	- " -	$13.6d'_{iF}+6.43d'_{i-1F}+6.33A'_{i-2(2)F}+0.20A'_{i-2(30)F}$	44.0	0.9961***
10	- " -	- " -	- " -	$14.4d'_{iF}+5.55d'_{i-1F}+7.24A'_{i-2(2)F}$	37.4	0.9945***
11	- " -	Tvarå	- " -	$14.6d'_{iF}+16.2d'_{i-1F}+14.4A'_{i-2(1)F}+1.86A'_{i-2(30)F}$	125	0.9906***
12	- " -	- " -	- " -	$9.41d'_{iF}+21.0d'_{i-1F}+7.16A'_{i-2(4)F}$	68.3	0.9879***

The radioecological sensitivity for pCi ^{137}Cs l^{-1} was 68.8 (Nos. 7, 9, 11), or 48.6 (Nos. 8, 10, 12).

Table C.3.4.1.a. Prediction models for ^{90}Sr in terrestrial animals (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Beef	Denmark[o)]	1963-75	$0.48d_i+0.07d_{i-1}+0.017A_{i-2(1)}+0.02A_{i-2(28)}$	1.37	0.9008***
2	- " -	- " -	- " -	$0.44d_i+0.10d_{i-1}+0.021A_{i-2(28)}$	1.38	0.9005***
3	Pork	- " -	- " -	$0.23d_i+0.13d_{i-1}+0.03A_{i-2(3)}+0.013A_{i-2(28)}$	1.01	0.8576***
4	- " -	- " -	- " -	$0.22d_i+0.14d_{i-1}+0.036A_{i-2(7)}$	0.72	0.8569***

o) The samples were collected in Copenhagen and were assumed to be representative of the country average.

Table C.3.4.1.b. Prediction models for ^{90}Sr in terrestrial animals (unit for radioecological sensitivity: pCi ^{90}Sr $(g\ Ca)^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $(g\ Ca)^{-1}_{(i)}$	Sensitivity	r
5	Mutton and lamb[x)]	Faroes	1961-76	$6.3d_{ip}+7.3d_{i-1p}+3.4A_{i-2(4)F}+0.1A_{i-2(28)F}$	15.6	0.6034**
6	- " -	- " -	- " -	$8.8d_{ip}+4.2d_{i-1p}+14A_{i-2(1)F}$	27.0	0.6198**
7	- " -	W.Greenland	1962-76[p)]	$20d_{iG}+3.4A_{i-1(7)G}$	52.7	0.8967**

p) The Greenland mutton sampling was incomplete, ^{90}Sr was thus not determined in 1963-1968 and in 1970, hence the prediction model is only a rough approximation and so is the sensitivity.

x) 1 kg mutton or lamb contained 0.1±0.04 g Ca (1SD; mean of 16 samples).

Table C.3.4.1.c. Prediction models for ^{90}Sr in terrestrial animals (unit for radioecological sensitivity: pCi ^{90}Sr $kg^{-1}y$ per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
8	Reindeer[s)]	W.Greenland	1962-75	$0.7d_{iG}+10.1d_{i-1G}+0.59A_{i-2(3)G}+0.046A_{i-2(28)G}$	14.9	0.9022***
9	- " -	- " -	- " -	$0.7d_{iG}+10.2d_{i-1G}+0.48A_{i-2(5)G}$	14.2	0.9031***
10	Musk-ox	E.Greenland	1964-73[a)]	$0.1d_{iG}+0.6A_{i-1(5)G}$	4	0.8078*

a) Musk-ox was not analyzed in 1966, 1969 and 1971, and the model is thus approximate.
s) 1 kg reindeer meat contained 0.12±0.07g Ca (1SD; mean of 56 samples).

Table C.3.4.2.a. Prediction models for ^{137}Cs in terrestrial animals (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Beef	Denmark[o)]	1963-75	$25.0d'_i+0.20d'_{i-1}+1.17A'_{i-2(2)}$	28.0	0.9929***
2	- " -	- " -	- " -	$24.7d'_i+1.88d'_{i-1}$	26.6	0.9906***
3	Pork	- " -	- " -	$16.6d'_i+16.4d'_{i-1}+1.17A'_{i-2(1)}$	34.2	0.9934***
4	- " -	- " -	- " -	$17.6d'_i+14.3d'_{i-1}+2.6d'_{i-2}$	34.5	0.9948***

o) Cf. note to Table C.3.4.1.

Table C.3.4.2.b. Prediction models for ^{137}Cs in terrestrial animals (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
5	Mutton and lamb[s]	Faroes	1961-76	$39d'_{iF}+0.16d'_{i-1F}+8.2A'_{i-2(3)F}+0.27A'_{i-2(30)F}$	82.2	0.8113***
6	- " -	- " -	- " -	$38d'_{iF}+1.3d'_{i-1F}+6.9A'_{i-2(4)F}$	76.0	0.8076***
7	- " -	W.Greenland	1961-76[p]	$27d'_{iG}+5.2A'_{i-1(12)G}+0.31A'_{i-1(30)G}$	128	0.6564*
8	- " -	- " -	- " -	$29d'_{iG}+4.9A'_{i-1(16)G}$	142	0.6584*

p) No samples from 1963, 1964, 1966, 1967, 1970.

s) 1 kg mutton or lamb contained 2.6±0.4 g K (1SD; mean of 35 samples).

Table C.3.4.2.c. Prediction models for ^{137}Cs in terrestrial animals (unit for radioecological sensitivity: pCi ^{137}Cs kg^{-1}y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi kg$^{-1}_{(i)}$	Sensitivity	r
9	Reindeer	W.Greenland	1962-75[q]	$157d'_{iG}+231d'_{i-1G}+200A'_{i-2(2)G}+21.7A'_{i-2(30)G}$	1799	0.6023*
10	- " -	- " -	- " -	$142d'_{iG}+254d'_{i-1G}+157A_{i-2(4)G}$	1226	0.6192*
11	Musk-ox	E.Greenland	1964-73[r]	$1.3d'_{iG}+8.0A'_{i-1(7)G}$	78	0.8450**

q) The 1968 sample was omitted in the prediction model because it was an outlier.

r) No samples from 1966 and 1969.

Table C.3.4.3. Prediction models for ^{90}Sr in bone from terrestrial animals (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Sheep bone	Faroes	1962-76	$12.5d_{iF}+5.2d_{i-1F}+6.4A_{i-2(2)F}+0.51A_{i-2(28)F}$	53.4	0.9185***
2	- " -	- " -	- " -	$12.9d_{iF}+4.1d_{i-1F}+8.6A_{i-2(2)F}$	37.9	0.9187***
3	- " -	W.Greenland	1961-75[a]	$3.9d_{iG}+3.3d_{i-1G}+2.6A_{i-2(10)G}+0.062A_{i-2(28)G}$	45.9	0.9374***
4	- " -	- " -	- " -	$3.6d_{iG}+2.8d_{i-1G}+2.93A_{i-2(9)G}$	43.0	0.9380***
5	Reindeer bone	- " -	1962-75[b]	$7.6d_{iG}+0.05d_{i-1G}+24.1A_{i-2(3)G}$	100	0.7733***

a) 1963-1964 and 1966-1970 were missing.

b) 1969 was missing.

Table C.3.5.1.a. Prediction models for ^{90}Sr in marine fish (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Cod	Denmark	1961-71[o]	$0.086d_i+0.005d_{i-1}+0.0064A_{i-2(2)}+0.024A_{i-2(28)}$	1.06	0.8856**
2	- " -	- " -	- " -	$0.086d_i+0.005d_{i-1}+0.029A_{i-2(15)}$	0.70	0.8843**
3	Cod and haddock	Faroes	1963-76	$0.024d_{iF}+0.20A_{i-1(2)F}+0.0046A_{i-1(28)F}$	0.69	0.7558**
4	- " -	- " -	- " -	$0.017d_{iF}+0.22A_{i-1(2)F}$	0.54	0.7553**

o) Except 1962, 1964, 1968, 1970.

Table C.3.5.1.b. Prediction models for ^{90}Sr in marine fish (unit for radioecological sensitivity: pCi ^{90}Sr kg^{-1}y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi kg$^{-1}_{(i)}$	Sensitivity	r
5	Plaice	Denmark	1963-71[p]	$0.056d_i+0.014d_{i-1}+0.014A_{i-2(28)}$	0.63	0.7350*
6	Salmon	W.Greenland	1963-74[q]	$0.97d_{iG}+0.046A_{i-1(28)G}$	3.8	0.6736*

p) Except 1966, 1970.

q) Except 1968, 1972.

Table C.3.5.2. Prediction models for ^{137}Cs in marine fish (unit for radioecological sensitivity: pCi $^{137}Cs\ kg^{-1}y$ per mCi $^{137}Cs\ km^{-2}$)

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Cod	Denmark	1963-71[a]	$2.3d'_i+0.0015d'_{i-1}+2.6A'_{i-2(5)}$	20	0.7941*
2	Cod and haddock	Faroes	1963-76	$0.36d'_{iF}+0.012d'_{i-1F}+0.096A'_{i-2(4)F}+0.027A'_{i-2(30)F}$	2.0	0.7281**
3	- " -	- " -	- " -	$0.35d'_{iF}+0.009d'_{i-1F}+0.12A'_{i-2(6)F}$	1.3	0.7183**
4	Cod	W.Greenland	1963-74[b]	$1.5d'_{iG}+0.86A'_{i-1(7)G}$	10	0.7825*
5	Plaice	Denmark	1963-71[c]	$0.34d'_i+0.86A'_{i-1(5)}$	6	0.2475
6	Salmon	W.Greenland	1963-74[d]	$1.83d'_{iG}+0.43d'_{i-1G}+0.37A'_{i-2(2)G}+0.19A'_{i-2(30)G}$	11.3	0.7999**
7	- " -	- " -	- " -	$1.80d'_{iG}+0.46d'_{i-1G}+0.43A'_{i-2(7)G}$	6.4	0.7963**

a) Except 1964, 1970.
b) Except 1965, 1966, 1968, 1970, 1972, 1973.
c) Except 1966, 1967.
d) Except 1968, 1972.

Table C.3.6.1. Prediction models for ^{90}Sr in sea mammals and hens' eggs (unit for radioecological sensitivity: pCi $^{90}Sr\ kg^{-1}y$ per mCi $^{90}Sr\ km^{-2}$)

No.	Sample	Area	Period	^{90}Sr-activity, pCi $kg^{-1}_{(i)}$	Sensitivity	r
1	Seal	Greenland[a]	1962-69	$0.145d_{iG}+0.024A_{i-1(4)G}+0.002A_{i-1(28)G}$	0.35	0.7740*
2	- " -	- " -	- " -	$0.146d_{iG}+0.024A_{i-1(5)G}$	0.31	0.7746*
3	Whale[d]	Faroes	1962-75[c]	$0.16d_{iF}+0.0036A_{i-1(28)F}$	0.30	0.8612***
4	- " -	Greenland[a]	- " -	$0.39d_{iG}+0.01A_{i-1(28)G}$	0.80	0.8409**
6	Hens' eggs	Denmark	1962-75[b]	$0.75d_i+0.08d_{i-1}+0.14A_{i-2(2)}+0.004A_{i-2(28)}$	1.33	0.8456***
7	- " -	- " -	- " -	$0.72d_i+0.19A_{i-2(2)}$	1.18	0.8471***

a) Godthåb precipitation data.
b) Except 1967, 1972.
c) Except 1964, 1967, 1971 and 1973.
d) The whales were from Faroese and Greenlandic waters.

Table C.3.6.2. Prediction models for ^{137}Cs in sea animals, sea birds and hens' eggs (unit for radioecological sensitivity: pCi ^{137}Cs $kg^{-1}{}_y$ per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi $kg^{-1}_{(1)}$	Sensitivity	r
1	Seal	Greenland[a)]	1963-69	$1.13d'_{iG}+0.08d'_{i-1G}+4.11A'_{i-2(1)G}$	5.3	0.7325*
2	Whale[d)]	Faroes	1962-75[b)]	$1.22d'_{iF}+1.29d'_{i-1F}+0.65A'_{i-2(1)F}+0.064A'_{i-2(30)F}$	5.9	0.9646***
3	- " -	- " -	- " -	$1.10d'_{iF}+1.58d'_{i-1F}+0.22A'_{i-2(5)F}$	4.2	0.9600***
4	- " -	Greenland[a)]	- " -	$2.1d'_{iG}+6.7d'_{i-1G}+0.24A'_{i-2(16)G}+0.01A'_{i-2(30)G}$	15	0.9862***
5	- " -	- " -	- " -	$2.1d'_{iG}+6.7d'_{i-1G}+0.25A'_{i-2(17)G}$	15	0.9862**
6	Hens' eggs	Denmark	1963-75[c)]	$0.001d'_{i}+1.63d'_{i-1}+0.031A'_{i-2(6)}$	1.90	0.9117***
7	- " -	- " -	- " -	$0.004d'_{i}+1.70d'_{i-1}$	1.70	0.9122***
8	Sea birds[g)]	Faroes	1962-75[f)]	$0.92d'_{iF}+0.23d'_{i-1F}+0.94A'_{i-2(2)F}$	3.4	0.5230
9	- " -	Greenland[a)]	- " -	$1.09d'_{iG}+0.46A'_{i-1(4)G}$	3.5	0.6017

a) Godthåb precipitation data.

b) Except 1964, 1967, 1971-1973.

c) Except 1967, 1972.

d) The whales were collected in Greenlandic and Faroese waters.

f) Except 1967, 1969, 1971-1974. The data were the calculated mean values obtained from a VAR-3. The models are tentative.

g) Brünnicks Guillemot, Puffin, Razorbill, Fulmar Petrel, Eider and Black Guillemot.

Table C.4.2.1. Prediction models for ^{90}Sr in Danish total diet (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	"Diet C"	8 zones	1961-76	$0.85d_i+1.50d_{i-1}+0.113A_{i-2(2)}+0.083A_{i-2(28)}$	6.00	0.9924***
2	- " -	- " -	- " -	$0.83d_i+1.52d_{i-1}+0.143A_{i-2(10)}$	4.34	0.9922***
3	- " -	Jutland	- " -	$0.87d_i+1.50d_{i-1}+0.150A_{i-2(2)}+0.082A_{i-2(28)}$	6.08	0.9903***
4	- " -	- " -	- " -	$0.85d_i+1.52d_{i-1}+0.169A_{i-2(8)}$	4.24	0.9900***
5	- " -	Islands	- " -	$0.81d_i+1.50d_{i-1}+0.056A_{i-2(2)}+0.091A_{i-2(28)}$	6.10	0.9915***
6	- " -	- " -	- " -	$0.80d_i+1.52d_{i-1}+0.118A_{i-2(15)}$	4.87	0.9915***
7	"Diet P"	Denmark	1959-76	$1.08d_i+1.57d_{i-1}+0.085A_{i-2(17)}+0.0065A_{i-2(28)}$	4.99	0.9883***
8	- " -	- " -	- " -	$1.08d_i+1.58d_{i-1}+0.090A_{i-2(18)}$	5.00	0.9884***

"Diet C" is the total diet measured as a mixed sample in June and December (D.4.2.3). "Diet P" is the total diet calculated from measurements of individual diet components; (dried milk from the 7 factories, grain from the 11 state experimental farms, potatoes from these farms, fruit and vegetables from the 8 zones, meat and eggs purchased in Copenhagen, fish from inner Danish waters, imported food purchased in Copenhagen and drinking water from the 8 zones).

Table C.4.2.2. Prediction models for ^{137}Cs in Danish total diet (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
1	"Diet C"	8 zones	1963-76	$1.56d_i^{\prime}+1.79d_{i-1}^{\prime}+0.88d_{i-2}^{\prime}$	4.22	0.9995***
2	- " -	Jutland	- " -	$1.24d_i^{\prime}+2.36d_{i-1}^{\prime}+0.77A_{i-2(1)}^{\prime}$	4.37	0.9971***
3	- " -	- " -	- " -	$1.52d_i^{\prime}+1.90d_{i-1}^{\prime}+0.91d_{i-2}^{\prime}$	4.33	0.9994***
4	- " -	Islands	- " -	$1.14d_i^{\prime}+2.23d_{i-1}^{\prime}+0.68A_{i-2(1)}^{\prime}$	4.06	0.9969***
5	- " -	- " -	- " -	$1.39d_i^{\prime}+1.82d_{i-1}^{\prime}+0.81d_{i-2}^{\prime}$	4.02	0.9995***
6	"Diet P"	Denmark	- " -	$1.70d_i^{\prime}+2.29d_{i-1}^{\prime}+0.131A_{i-2(1)}^{\prime}+0.0111A_{i-2(30)}^{\prime}$	4.66	0.9994***
7	- " -	- " -	- " -	$1.68d_i^{\prime}+2.32d_{i-1}^{\prime}+0.050A_{i-2(4)}^{\prime}$	4.29	0.9993***
8	- " -	- " -	- " -	$1.79d_i^{\prime}+2.17d_{i-1}^{\prime}+0.23d_{i-2}^{\prime}$	4.19	0.9994***

(Cf. the note to Table C.4.2.1)

Table C.4.3.1.a. Prediction models for ^{90}Sr in human vertebrae in Denmark (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Newborn: 0-30 d	Zones I,II,III, IV,VI	1960-76	$0.081d_i+0.124d_{i-1}+0.023A_{i-2(1)}+0.020A_{i-2(28)}$	1.03	0.9219***
2	- " -	- " -	- " -	$0.075d_i+0.137d_{i-1}+0.021A_{i-2(28)}$	1.06	0.9193***
3	Infants: 4-12 mo.	- " -	1963-76	$0.157d_i+0.35d_{i-1}+0.113A_{i-2(1)}+0.028A_{i-2(28)}$	1.74	0.9638***
4	- " -	- " -	- " -	$0.139d_i+0.38d_{i-1}+0.060A_{i-2(7)}$	1.10	0.9594***
5	Infants: 13-60 mo.	- " -	1963-76	$0.055d_i+0.32d_{i-1}+0.18A_{i-2(1)}+0.020A_{i-2(28)}$	1.35	0.9480***
6	- " -	- " -	- " -	$0.086d_i+0.29d_{i-1}+0.34A_{i-2(1)}$	0.72	0.9347***

Table C.4.3.1.b. Prediction models for ^{90}Sr in human vertebrae in Denmark and the Faroes (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
7	Children: 6-12 y	Zones I,II,III, IV,VI	1962-76	$0.132d_{i-1}+0.071A_{i-2(2)}+0.022A_{i-2(28)}$	1.18	0.9226***
8	- " -	- " -	- " -	$0.138d_{i-1}+0.065A_{i-2(6)}$	0.67	0.9201***
9	Adolescents: 13-19 y	- " -	1962-76	$0.165d_{i-1}+0.145A_{i-2(1)}+0.022A_{i-2(28)}$	1.19	0.9713***
10	- " -	- " -	- " -	$0.187d_{i-1}+0.058A_{i-2(6)}$	0.66	0.9589***
11	Adults: 20-29 y	- " -	1960-76	$0.086d_{i-1}+0.111A_{i-2(1)}+0.023A_{i-2(28)}$	1.12	0.9575***
12	- " -	- " -	- " -	$0.102d_{i-1}+0.051A_{i-2(7)}$	0.59	0.9464***
13	Adults: > 29 y	- " -	1960-76	$0.060d_{i-1}+0.083A_{i-2(1)}+0.023A_{i-2(28)}$	1.05	0.9398***
14	- " -	- " -	- " -	$0.075d_{i-1}+0.041A_{i-2(9)}$	0.59	0.8997***
15	Newborn: 0-30 d	Faroes	1965-76	$0.148d_{iF}+0.113d_{i-1F}+0.054A_{i-2(2)F}$	0.39	0.7700**
16	- " -	- " -	- " -	$0.25d_{iF}+0.063A_{i-1(1)F}+0.0134A_{i-1(28)F}$	0.85	0.7553**

Table C.4.4.1. Prediction Models for ^{90}Sr in shed deciduous teeth and infant bone (unit for radioecological sensitivity: pCi ^{90}Sr (g Ca)$^{-1}$y per mCi ^{90}Sr km^{-2})

No.	Sample	Area	Period[a]	^{90}Sr-activity, pCi (g Ca)$^{-1}_{(i)}$	Sensitivity	r
1	Teeth[b,c]	Denmark	1951-70	$0.116d_i+0.187d_{i-1}+0.108A_{i-2(4)}+0.0004A_{i-2(28)}$	0.89	0.9926***
2	- " -	- " -	- " -	$0.187d_i+0.116d_{i-1}+0.109A_{i-2(4)}$	0.88	0.9926***
3	- " -	Faroes	1951-67	$0.264d_i+0.127d_{i-1}+0.43A_{i-2(1)}+0.0076A_{i-2(28)}$	1.12	0.9540***
4	- " -	- " -	- " -	$0.168d_i+0.23d_{i-1}+0.23A_{i-2(2)}$	0.95	0.9585***
5	Teeth and infant bone[c]	Denmark	1951-76	$0.119d_i+0.26d_{i-1}+0.123A_{i-2(1)}+0.032A_{i-2(28)}$	1.78	0.9825***
6	- " -	- " -	- " -	$0.105d_i+0.29d_{i-1}+0.067A_{i-2(7)}$	1.04	0.9803***

a) The year (i) is the year when the tooth donor was 1 yr old, i.e. the birth year of the donor is i-1.

b) As the first years were under-represented with regard to incisors, and the opposite was the case for the latest part of the period, the VAR-3 figures have been used instead of the original data (cf. B.4). As explained in (XII), the tooth-data from donors born prior to 1957 were corrected for missing creta praeparata in the diet, before they were used in the calculation of the prediction models.

c) The values were estimated from VAR-3 (cf. B.4). The bones in No. 5 were from 4-12 months old infants.

Table C.4.5.1. Prediction models for ^{137}Cs in adults working at Risø (unit for radioecological sensitivity: pCi ^{137}Cs (g K)$^{-1}$y per mCi ^{137}Cs km^{-2})

No.	Sample	Area	Period	^{137}Cs-activity, pCi (g K)$^{-1}_{(i)}$	Sensitivity	r
1	Both sexes	Zealand	1963-76	$1.28d'_i+5.66d'_{i-1}+4.81A'_{i-2(1)}$	11.8	0.9954***
2	- " -	- " -	- " -	$2.54d'_i+3.79d'_{i-1}+4.87d'_{i-2}$	11.2	0.9881***
3	Males	- " -	- " -	$0.81d'_i+6.88d'_{i-1}+5.42A'_{i-2(1)}$	13.1	0.9969***
4	- " -	- " -	- " -	$2.16d'_i+4.94d'_{i-1}+5.36d'_{i-2}$	12.5	0.9857***
5	Females	- " -	- " -	$1.74d'_i+4.56d'_{i-1}+3.93A'_{i-2(1)}$	10.2	0.9943***
6	- " -	- " -	- " -	$2.72d'_i+3.13d'_{i-1}+3.90d'_{i-2}$	9.8	0.9876***

APPENDIX D

NOTES TO THE MAIN TEXT

This appendix comprises details considered as a useful supplement to the main text. The numbering corresponds to that of the corresponding passages in the main text.

D.1.2. Summary of the history of nuclear weapons testing

The Research Institute of National Defence in Sweden (FOA) has collected the information available on nuclear explosions for the period 1945-1972 (Za73). The first thermonuclear explosion took place on 31 October, 1952, on the Enewetak atolls ($\sim 11^{\circ}$N, $\sim 162^{\circ}$E) in the Pacific Ocean. This test initiated global or worldwide fallout. Before that time the non-local fallout had been limited to rather narrow bands around the latitudes of the test sites; such fallout has been called tropospheric or intermediate fallout. In November 1955 the USSR performed their first thermonuclear test at Novaya Zemlya ($\sim 70^{\circ}$N, $\sim 50^{\circ}$E). Until the end of 1958, USA, USSR and UK had tested fusion weapons with a total fission yield of approx. 92 megatons TNT (Fe63). Of this yield, 38 Mt were air explosions, and 54 Mt were surface shots. In 1959-1960 there was a temporary test moratorium, but in September 1961 the USSR and USA resumed their atmospheric tests. During 1961-1962 101 Mt TNT*) fission yield (Fe63) were detonated as air explosions. Since then none of the three original nuclear powers has performed atmospheric tests.

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*) According to the inventory estimates made by UNSCEAR (Un77) this fission yield estimate seems too high as the ^{90}Sr production in the 1961-1962 series was only approx. 9 MCi corresponding to a fission yield of approx. 83 Mt TNT.

In 1967 the first Chinese thermonuclear weapon test took place at Lop Nor in the Sinkiang Province (∿ 40°N, ∿ 90°E), and in the following year France detonated a thermonuclear device in the South Pacific (∿ 20°S, ∿ 140°W). China has carried out thermonuclear shots in the atmosphere in 1967, 1968, 1969, 1970, 1973, 1974 and 1976. These explosions have generally been of the order of 1-2 Mt TNT fission resulting in a global deposition every post-test year of approx. 0.1 - 0.2 MCi ^{90}Sr (Te77). The French test series was only of minor importance as a source of fallout in the Northern hemisphere as the inter-hemispheric exchange of air masses is so slow that most fallout is deposited on the hemisphere of testing (Un77), and also because the total fission yield was only of the order of 5 Mt TNT (Za73).

D.1.2.1. Local variations in the concentrations of ^{90}Sr and ^{137}Cs in air samples

During the period 1962-1974, 3 stations from the HASL (Ha58-78) programme: Thule, Greenland (76° 36' N, 68° 35' W), Moonsonee, Canada (51° 16' N, 80° 30' W) and Sterling, Virginia (38° 58' N, 77° 25' W), a Harwell station (Ae58-74): Chilton, U.K. (51° 30' N, 05° W), and Risø (55° 40' N, 12° 08' E) were compared. The relative ^{90}Sr air concentrations for the period were obtained from a VAR-3 anova (cf.B.3): Thule: 0.99, Moonsonee: 1.05, Sterling: 1.28, Chilton: 0.85, and Risø: 0.83. The anova showed that the variation between stations was significant. This was to be expected over a latitude band of nearly 40°. An anova of the data from Chilton, Moonsonee and Risø, which are all within a 10° latitude zone, showed that Chilton and Risø did not differ significantly, but that Moonsonee was significantly higher. It was anticipated that the annual ^{90}Sr air concentrations at Risø were representative of the annual means of the air levels for the entire country. This assumption was reasonable also in view of the negligible local variations in rainwater concentrations of ^{90}Sr measured in the various parts of the country (cf.1.3.1.).

The local variation of the $^{137}Cs/^{90}Sr$ ratio was also studied at the five locations mentioned above. During 1962-1974 the

annual mean ratios were 1.67 ± 0.09 (1 SE) at Risø, 1.72 ± 0.09 at Chilton, and 1.57 ± 0.07 at the 3 HASL stations (Thule, Moonsonee and Sterling). An anova of the annual mean $^{137}Cs/^{90}Sr$ ratios from the five locations showed no significant variation between locations nor between years. Hence, the ^{137}Cs air concentrations followed those of ^{90}Sr both with time and location.

Table D.1.2.1. Total deposition of ^{90}Sr in the northern hemisphere related to ^{90}Sr and ^{137}Cs concentrations in ground level air at Risø.

Year	MCi ^{90}Sr deposited (1) (Un 77)	fCi ^{90}Sr m^{-3} (2) (RRD 59-76)	fCi ^{137}Cs m^{-3} (3) (RRD 59-76)	(2)/(1) (4)	(3)/(1) (5)
Pre 1958	1.80	(17.5)			
1958	0.63	5.0		7.94	
1959	1.05	10.5		10.0	
1960	0.26	2.1	pre 1962 (67.4)	8.08	
1961	0.35	2.0		5.71	
1962	1.44	10.6	23.1	7.36	16.04
1963	2.62	30.5	65.6	11.64	25.04
1964	1.66	21.7	31.2	13.07	18.80
1965	0.77	6.7	10.6	8.70	13.77
1966	0.33	2.94	5.7	8.91	17.27
1967	0.17	1.10	2.14	6.47	12.59
1968	0.20	1.42	2.38	7.10	11.90
1969	0.15	1.41	2.45	9.40	16.33
1970	0.21	2.16	3.40	10.29	16.19
1971	0.19	1.97	2.67	10.37	14.05
1972	0.09	0.84	1.37	9.33	15.22
1973	0.03	0.40	0.47	13.33	15.67
1974	0.12	1.42	1.96	11.83	16.33
1975	0.06	0.90	1.30	15.00	21.67
		Σ 121 fCi y m^{-3}	Σ 222 fCi y m^{-3}	$\bar{x}$ 9.7 SD 2.5	$\bar{x}$ 16.5 SD 3.5

The figures in brackets are the integrated air concentrations (of ^{90}Sr and ^{137}Cs before Danish measurements began) estimated from the mean ratios found in (4) and (5), respectively.

Table D.1.3.2. Estimates of ^{90}Sr deposition in Denmark before Danish measurements began (unit: mCi ^{90}Sr km^{-2} y^{-1})

Year	Estimate 1 (XI)	Estimate 2 (X)	Estimate 3
1950	0.02	0.02	0.02
1951	0.1	0.1	0.1
1952	0.1	0.2	0.2
1953	0.4	0.5	0.5
1954	0.8	1.0	1.9
1955	1.7	2.0	2.5
1956	1.8	2.0	3.1
1957	2.0	2.1	3.1
1958	3.4	4.4	4.3
1959		4.7	6.1

In Estimates 1 and 2 the fallout rates during 1950-1954 were assessed from information on nuclear test explosions during these years (Ha 73, Fe 63). Since 1955, Estimate 1 has been based on the assumption that the ^{90}Sr concentrations in Danish precipitation equalled those in rainwater from Milford Haven in the UK (As 58-74). Estimate 2 assumed that the mean ratio between the fallout rates in Denmark and in Milford Haven during 1955-1958 equalled that observed during 1962-1967, i.e. 0.82. Estimate 3 used the same method as 1 and 2 for 1950-1953, but from 1954 the estimates were calculated from the HASL data (Ha 58-78) observed in New York, considering that the mean ratio between ^{90}Sr fallout in Denmark and New York was 0.7 during the period 1962-1976. Estimate 3 was preferred to the others because the accumulated fallout actually measured in the soil samples collected at the beginning of the sixties agreed with the values obtained from this estimate. Moreover, the ^{90}Sr air data also showed a better agreement with this estimate, as was suggested from the agreement between the infinite integrated deposition data based upon air and precipitation measurements (cf. 1.3.2.).

D.1.3.3. Calculation of the fallout-weighted mean precipitation at the state experimental farms (Fig.A.1.1.3.1 and table A.1.1.6.2.)

For 1953-1961 the amounts of precipitation were assessed from the meteorological measurements at the stations (Lu71, Me59-77). The mean for the period was 664 mm y^{-1}. If the individual years were weighted by the ^{90}Sr fallout rates measured in 1962-1974, this weighted mean became 638 mm y^{-1}.

For 1962-1974 fallout rain-bottles were used for the estimation of precipitation; the unweighted mean was 638 mm, and the fallout weighted mean was also 638 mm. As the meteorological observations yielded 5% higher amounts of precipitation than the rain-bottles when the two systems were compared (RRD59-76), the precipitation estimate for the period 1953-1961 should be divided by 1.05 to obtain the overall weighted mean of precipitation (in rain-bottles):

$$1953\text{-}1974: \frac{((638:1.05) \cdot 24.1) + (638 \cdot 48.6)}{72.7} = 628 \text{ mm } y^{-1}.$$

where 24.1 and 48.6 were the total depositions of ^{90}Sr in 1953-1961 and in 1962-1974, respectively, and 72.7 was the deposition of the entire period.

D.1.4.1. An estimate of the transfer of ^{90}Sr from Danish ground water to the biosphere

The amount of ^{90}Sr returning to the biosphere as a result of the pumping out of ground water may be estimated if it is recalled that the amount of ground water pumped out annually in Denmark is $7 \cdot 10^{11}$ l y^{-1} (Be71), or $1.63 \cdot 10^{7}$ l km^{-2} y^{-1}. 1 Ci ^{90}Sr deposited per km^{-2} in Denmark yields $7.4 \cdot 10^{-12}$ Ci ^{90}Sr l^{-1} y in the ground water (table C.1.4.1. No. 1), hence $7.4 \cdot 10^{-12} \cdot 1.63 \cdot 10^{7} = 121 \cdot 10^{-6}$ of the ^{90}Sr deposited returns to the biosphere. Thus, for each curie of ^{90}Sr deposited as fallout on the terrestrial surface of Denmark, 121 μCi return to the biosphere as a result of the pumping out of ground water. The total amount of ^{90}Sr returning to the biosphere is greater than this because streams also carry ground water. The theoretical maximum transfer of ^{90}Sr from Danish ground water to the biosphere may be estimated from the annual contribution of precipitation to ground water being $4.7 \cdot 10^{12}$ l y^{-1} (Be71), or $1.1 \cdot 10^{8}$ l km^{-2} y^{-1}. Thus: $7.4 \cdot 10^{-12} \cdot 1.1 \cdot 10^{8} = 814 \cdot 10^{-6}$, or of the order of 1‰ of the deposited ^{90}Sr may return to the biosphere if the Danish ground water shows the same ^{90}Sr mean level as that of the eight borings.

D.1.4.2. Estimate of ^{90}Sr run-off with Danish streams

According to table C.1.4.1. No. 3, the instant run-off of ^{90}Sr from nuclear weapons testing until 1974 was:

$$0.17 \cdot 73 = 12.4 \text{ pCi } ^{90}Sr \text{ } l^{-1} \text{ y}$$

and the infinite time-integrated delayed run-off was:

$$73 \cdot (0.0028 \cdot 8.2 + 0.0035 \cdot 39.9) = 12 \text{ pCi } ^{90}Sr \text{ } l^{-1} \text{ y}$$

Of this amount:

$$(0.0028 \cdot 8.2 \cdot 20.1 + 0.0035 \cdot 39.9 \cdot 53.8) = 8 \text{ pCi } ^{90}Sr \text{ } l^{-1} \text{ y}$$

still remain to be released as delayed run-off (the ^{90}Sr accumulated by 1973 in Danish soils, assuming effective halflives of 6* and 28* years for ^{90}Sr, was 20.1 mCi km^{-2} and 53.8 mCi km^{-2}, respectively). In 1950-1974 the amount of delayed run-off was:

$$12 - 8 = 4 \text{ pCi } ^{90}Sr \text{ } l^{-1} \text{ y},$$

and the total run-off during 1950-1974 was:

$$12.4 + 4 = 16.4 \text{ pCi } ^{90}Sr \text{ } l^{-1} \text{ y}.$$

As the annual run-off is approx. $12 \cdot 10^{12}$ l (Je69) and the area of Denmark is 43,069 km^2, the run-off during 1950-1974 corresponded to:

$$\frac{16.4 \cdot 10^{-12} \cdot 12 \cdot 10^{12} \cdot 10^{3}}{43{,}069} = 4.6 \text{ mCi } ^{90}Sr \text{ } km^{-2}.$$

The time-integrated soil level during 1950-1974 was 836.6 mCi ^{90}Sr km^{-2} y, hence the annual mean removal was 5 o/oo. The total infinite time run-off in Denmark was:

$$(12.4 + 12) \cdot 12 = 293 \text{ Ci } ^{90}Sr$$

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*The time integrated levels were calculated from $\frac{e^{-\mu}}{1 - e^{-\mu}}$ (cf C.2) where μ is the decay constant. For 6 year the expression is 8.2 and for 28 year: 39.9.

corresponding to 6.8 mCi ^{90}Sr km^{-2} or 9.3% of the total ^{90}Sr deposited (= 73 mCi km^{-2}), and the run-off remaining to be released as delayed run-off in future years from 1975 and then onwards was 6.8 - 4.6 = 2.2 mCi ^{90}Sr km^{-2}.

Table D.1.5.1. The ^{90}Sr and ^{137}Cs concentrations in Danish waters related to salinity

Period	Unit	Equation		No.
1967-1971	pCi ^{90}Sr l^{-1}	= 0.94 - 0.018	salinity o/oo	1
1972	- " -	= 0.97 - 0.020	- " -	2
1973	- " -	= 0.95 - 0.014	- " -	3
1974	- " -	= 0.93 - 0.010	- " -	4
1975	- " -	= 0.79 - 0.006	- " -	5
1976	- " -	= 0.71 - 0.002	- " -	6
1972	pCi ^{137}Cs l^{-1}	= 0.80 - 0.0043	- " -	7
1973	- " -	= 0.60 + 0.012	- " -	8
1974	- " -	= 0.54 + 0.018	- " -	9
1975	- " -	= 0.64 + 0.010	- " -	10
1976	- " -	= 0.53 + 0.019	- " -	11

Eqs. 1, 2, 4, 8, 9, 10 and 11 all showed significant or probably significant regression. The other equations did not show any regression.

Table D.1.5.2. Strontium-90 in deep sea water collected by the Dana in the North Atlantic

Location		Year	Unit	Depth					
				500 m	1000 m	1500 m	2000 m	2500 m	3500 m
38°54'N	23°06'W	1966	pCi ^{90}Sr l^{-1}	0.046	0.011		0.005		0.004
			Salinity o/oo	35.4	35.6		35.1		34.8
42°29'N	13°32'W	1966	pCi ^{90}Sr l^{-1}	0.038	0.027		0 020		0.018
			Salinity o/oo	35.7	36.9		35.0		34.8
62°50'N	34°20'W	1966	pCi ^{90}Sr l^{-1}	0.093	0.019	0.048	0.040		
			Salinity o/oo	-	-	-	-		
63°18'N	3°50'W	1967	pCi ^{90}Sr l^{-1}	0.080				0.079	
			Salinity o/oo	34.9				34.9	
60°49'N	4°15'W	1971	pCi ^{90}Sr l^{-1}		0.020				
			Salinity o/oo		33.6				
62°00'N	32°20'W	1971	pCi ^{90}Sr l^{-1}		0.061			0.030	
			Salinity o/oo		33.8			33.4	
68°19'N	56°52'W	1971	pCi ^{90}Sr l^{-1}	0.103					
			Salinity o/oo	33.2					
62°N	35°25'W	1973	pCi ^{90}Sr l^{-1}		0.083			0.044	
			Salinity o/oo		-			35.8	

D.1.6.1.1. Removal processes for radionuclides deposited on the surface of the earth. Besides radioactive decay, other processes influence the removal of radionuclides from the soil, and this explains why the observed halflife may be lower than the radiological halflife. Five processes may be mentioned:

1. Run-off, which includes physical removal of soil by running water as well as chemical extraction of radionuclides fixed to the soil minerals.

2. Removal by crops, i.e. removal of the activity taken up by the crops (by direct or indirect contamination) when the crops are harvested or eaten in the fields by livestock.

3. Soil contamination of crops. Soil and thus radioactive debris may adher to crops when, for example, root crops are removed from the fields.

4. Mechanical removal of soil, i.e. removal by means of agricultural machinery (e.g. tractor wheels).

5. Soil drift, i.e. wind erosion, which is of special importance for light soils before they are covered by vegetation, i.e. in early spring.

The importance of processes 1 and 2 depends upon the fallout rate as well as on the accumulated fallout, while 3, 4, and 5 primarily depend upon the accumulated fallout.

1. Run-off

According to 1.4.2., the total run-off during 1950-1974 corresponded to 4.6 mCi $^{90}Sr\ km^{-2}$, 3.5 mCi $^{90}Sr\ km^{-2}$ were so-called instant run-off, i.e. dependent upon the fallout rate, and 1.1 mCi km^{-2} were delayed run-off dependent upon the accumulated ^{90}Sr in the soil. If the 4.6 mCi $^{90}Sr\ km^{-2}$ were related to the time-integrated soil level during 1950-1974 (836.6 mCi $^{90}Sr\ km^{-2}$ y), it appeared that approx. 5% p.a. of the ^{90}Sr in Danish soils had been removed by run-off. As the fallout rate decreases, the importance of instant fallout declines, and for future years a delayed run-off of 2.2 mCi $^{90}Sr\ km^{-2}$ is expected (cf.D.1.4.2.), which, compared to the infinite time-integrated ^{90}Sr level in the soil (2127 mCi $^{90}Sr\ km^{-2}$ y), corresponds to an annual run-off of approx. 1% p.a. During 1950-1974 a mean decay factor of 0.005 y^{-1} may be applied and from then onwards the decay factor is estimated at 0.001 y^{-1}.

2. Removal by crops

During 1950-1974, 4.9 mCi $^{90}Sr\ km^{-2}$ and 3.4 mCi $^{137}Cs\ km^{-2}$ were removed from arable land by crops (table D.1.6.1.1.). However, 80% returned in the form of manure to the fields, and the net removal was thus 1.0 mCi $^{90}Sr\ km^{-2}$ and 0.7 mCi $^{137}Cs\ km^{-2}$, corresponding to approx. 1% p.a. of the accumulated activity in the soil. From the prediction models for various crops it

appeared that direct fallout contributed approx. 3/4 of the ^{90}Sr activity in the crops during 1950-1974 and nearly all of the ^{137}Cs. Provided the fallout rate continues to decrease, we may expect a decreasing removal of ^{90}Sr and ^{137}Cs by crops in the coming years. From the prediction models it was estimated that the future total net removal of ^{90}Sr from arable land (but not necessarily from the terrestial environment as a whole) will be approx. 0.3 mCi km^{-2}, or 0.1 ‰ p.a. For the period 1950-1974 the mean decay factor was estimated at 0.001 y^{-1} and thereafter at 0.0001 y^{-1}, i.e. negligible.

Table D.1.6.1.1. An estimate of the total ^{90}Sr and ^{137}Cs uptake by Danish crops 1950-1974.

Sample	Mean area km^2	Mean production kg y^{-1}	^{90}Sr pCi kg^{-1} y	^{90}Sr Total Ci	^{137}Cs pCi kg^{-1} y	^{137}Cs Total Ci
Barley-grain [a]	14.000	5×10^9	1.48×10^3	7.4	3.74×10^3	18.7
Straw		2×10^9	14.15×10^3	28.3	7.35×10^3	14.7
Roots	5.000	20×10^9	0.93×10^3	18.6	0.09×10^3	1.9
Leaves		5×10^9	3.00×10^3	15.0	1.22×10^3	6.1
Grass	6.000	20×10^9	2.89×10^3	57.9	2.30×10^3	46.0
Permanent grass	4.000	13×10^9	2.89×10^3	(37.6)	2.30×10^3	(29.9)
Other crops	1.000	3×10^7	$\sim3\times10^3$	0.1	$\sim1.22\times10^3$	$\sim$ 0
Σ	30.000			127.3		87.4

The time integrals for the period 1950-1974 were calculated as the difference between the infinite time integrals since 1950 and the infinite time integrals since 1974 applying the pertinent prediction models: barley grain: Table C.2.2.3 Nos. 3 and 4, Table C.2.2.6 No. 3; straw, roots and leaves: Table C.2.4.3 and Table C.2.4.4; grass: Table C.2.4.1 No. 3 and Table C.2.4.2 No. 1; other crops: as for leaves.

Total fallout over 30.000 - 4.000 = 26.000 km^{-2}: 26×73 Ci = 1898 Ci ^{90}Sr and 1.6×1898 = 3037 Ci ^{137}Cs. Crops carried 6.7% ^{90}Sr and 2.9% ^{137}Cs. However, 80% returns as manure to the fields, hence 1.3% ^{90}Sr and 0.6% ^{137}Cs were permanently removed by the crops.

a) All grain was regarded as barley.

3, 4, and 5 - various soil removal processes

The last three processes are all tantamount to a physical removal of soil from the fields. However, quantitative data on soil drift in particular and on the mechanical removal of soil are not at hand. Some information on soil contamination

of root crops was available. Approx. 5% p.a. of the ploughing layer was removed from sugar-beet fields with clay soils in Lolland (RRD75) during 1950-1974. From the time-integrated ^{90}Sr level in the ploughing layer, the ^{90}Sr removal during 1950-1974 was estimated at approx. 3 mCi ^{90}Sr km^{-2}, or approx. 4% p.a. of the total ^{90}Sr activity in the soil. As soil drift is more important for sandy soils than for clay soils, and as soil removal by crops and machinery, on the other hand, is less important for sandy soils, it may be assumed as a provisional estimate that the average removal of ^{90}Sr from Danish soils by processes 3, 4 and 5 during 1950-1974 was 4 mCi ^{90}Sr km^{-2}, or 5% p.a. of the ^{90}Sr in the soil. As the removal of ^{90}Sr by the three processes primarily depends upon the ^{90}Sr content of the ploughing layer, we may use the infinite time integral of the ^{90}Sr in the ploughing layer to estimate the future removal of ^{90}Sr by these processes at approx. 2 mCi ^{90}Sr km^{-2}, or 1% p.a. of the future ^{90}Sr content of the soil. The mean decay factor becomes 0.005 y^{-1} for the period 1950-1974 and 0.001 y^{-1} thereafter.

The overall mean decay factors were calculated from the above estimates: For the period 1950-1974 $\lambda_c = 0.025 + 0.005 + 0.001 + 0.005 = 0.036$ y^{-1} and for future years $\lambda_c = 0.025 + 0.001 + 0 + 0.001 = 0.0027$ y^{-1}. The corresponding halflives were 19 y and 26 y, respectively, being compatible with the ^{90}Sr levels found in cultivated soils. Although the estimates of the decay factors for the various non-radioactive removal processes are thus uncertain, and although the decay factors change with time, these factors are not seriously underestimated; the observed halflife of ^{90}Sr in cultivated soil was about 20 years during 1950-1974. In the coming years the observed halflives of ^{90}Sr in soil will approach the radiological halflives, provided that the fallout rate continues to decrease.

Table D.1.6.1.2. Strontium-90 and Cesium-137 in Greenlandic and Faroese soils.

Location Latitude Longitude (sampling year)	Sample depth interval in cm	pCi ^{90}Sr kg^{-1}	mCi ^{90}Sr km^{-2}	pCi ^{137}Cs kg^{-1}	mCi ^{137}Cs km^{-2}	Annual mean precipitation in cm (1962-1975)
Dundas	0-20	61±2	11.7±0.4	110	21	
76°34'N						~ 150
68°50'W						
(1970)	20-35	13±1	2.0±0.2	31	5	(Hard 72)
Godthåb	0-20	141±6	33.3±1.4	355	84	
64°N						782
52°W						
(1970)	20-35	29±1	5.0±0.2	12	2	(Da 63-76)
Thorshavn	0-20	768	49.7	2891	190	
62°02'N						
6°45'W	20-30	266±33	12.6±1.5	316	15	1377
(1967)	30-50	141	12.9	171	16	(Du 75)
Tvørå	0-20	1310	91.7	3051	217	
61°33'N						
6°41'W	20-30	60	1.8	129	4	(~1500)
(1967)	30-50	37	4.8	89	12	

D.1.6.2. Estimate of the effective halflife or ^{90}Sr in Danish soils

The vertical distribution of ^{90}Sr has implications for the direct contamination of crops. ANDERSEN (An73) conducted a study of the uptake by barley, rye-grass, red clover and root crops of ^{90}Sr placed at various depths in the soil profile of five different Danish soils. The uptake differed for the various species and soil types. If, however, a regression analysis was applied to the data, the relative root uptake of ^{90}Sr by the crops followed with approximation the equation

$$U = e^{-c\sqrt{a_m}} \qquad \text{(Eq.D.1.6.2.1)}$$

where c is the "relative root uptake factor", which varied between 0.1 and 0.5 ($cm^{-0.5}$). In a given soil the highest c-values were found for grass, the lowest for barley and roots. Sandy soils generally showed lower c-values than clayish soils. As shown by several authors (Ru66a), numerous factors influence the root uptake of ^{90}Sr; the present model should consequently be considered as an approximation only.

From Eqs.1.6.1 and 1.6.2 the "fallout-weighted median depth" a_{m_n} by the end of the year n was obtained:

$$a_{m_n} = \frac{a_{m_o} \sum_{i=0}^{i=n} \sqrt{n + 1 - i}\, d_i e^{-(n - i + 1)\lambda_r}}{An} \qquad \text{(Eq.D.1.6.2.2)}$$

where a_{m_o} was the median depth calculated from Eqs.1.6.4 and 1.6.2. for t = 1, i.e. for n = 0 in Eq.D.1.6.2.2. A_n was the accumulated fallout (Eq.1.6.1.). If the calculated a_{m_n} from Eq.1.6.2.2. was inserted into Eq.D.1.6.2.1., the relative root uptake of a year n: U_n was obtained. The calculated U_n values for various a and c values for each year in the period 1950-1975 were then by correlation analysis compared with the cumulated ^{90}Sr from fallout, assuming effective half-lives of the soil

Table D.1.6.2.1. Calculated effective half-lives for the root uptake of ^{90}Sr from uncultivated soils.

Period	a = 1			a = 2			a = 3		
	c = 0.1	c = 0.2	c = 0.4	c = 0.1	c = 0.2	c = 0.4	c = 0.1	c = 0.2	c = 0.4
1950-1964	19	14	10	20	15	7	21	16	11
1965-1975				18		7			
1976-1989	20	16	12	20	17	12	21	18	14
1990-2000				23		14			

a is the "vertical distribution coefficient" (cf. Eq. 1.6.3.).
c is the "relative root uptake factor" (Eq. 1.6.5.).
The half-lives (given in years) were calculated as described in 1.6.2. In the calculations of future half-lives it was assumed that no fresh ^{90}Sr was injected into the atmosphere after 1975, and that the effective half-life of the ^{90}Sr in the stratosphere was 10 months (cf. 1.2.). In fact, ^{90}Sr has already been injected into the atmosphere since 1975, but this does not change the tendency of the figures in the table.

^{90}Sr varying from 1 to 28 years (table D.1.6.2.1.). The future situation until year 2000 was also considered. It appeared that the effective half-life for the indirect contamination of crops will increase in the future. The parametric study showed that variations in the "vertical distribution coefficient" a

were of less importance for root uptake than variations in the "relative root uptake factor" c. An effective half-life of the order of 10 years may be expected for a large median depth combined with a crop with a shallow root system. A longer effective half-life may, on the other hand, be found in a soil with a shallow median depth for a crop with a low "relative root uptake factor".

As regards the vertical distribution in cultivated soil, it was presumed that the ^{90}Sr activity of the ploughing layer, which is approx. 20 cm thick, was uniformly distributed. The effective half-life of the ^{90}Sr in the ploughing layer was estimated from the measurements in 1962 (RRD62) and 1975 (RRD75) at 9 years ($\lambda_1 = 0.077\ y^{-1}$). Below the ploughing layer, the situation was assumed to be analogous to the conditions in undisturbed soil, i.e. the vertical activity distribution was described by (Eq.1.6.3.) and the penetration of the activity front of a given year's deposit followed 10 $\sqrt{t'}$, where t' was the time in years since the activity entered the layer below the ploughing layer. If the cultivated soil was ploughed every year, it was assumed that the activity introduced into the ploughing layer during a year (i) started to enter the soil below the ploughing layer in the following year (i+1). The effective half-life of the ^{90}Sr in the ploughing layer was, in addition to the above-mentioned removal processes (D.1.6.1.1.), also influenced by the vertical transfer of activity to the soil below the ploughing layer. The decay factor λ_p for this depletion of ^{90}Sr was given as:

$$\lambda_p = \lambda_1 - \lambda_c = 0.077 - 0.036 \sim 0.04\ y^{-1}$$

corresponding to an annual downward transfer of 4% of the ^{90}Sr present in the ploughing layer.

Analogous to Eq.1.6.1., we now find for cultivated soils:

$$A_{cn} = \sum_{i=0}^{i=n} d_i e^{-\lambda_c (n - i + 1)} \qquad \text{(Eq.D.1.6.2.3)}$$

where A_{cn} is the accumulated ^{90}Sr in mCi km^{-2} in cultivated soil as a whole n years after the deposition began (i.e. in 1950), d_i is the fallout rate in mCi ^{90}Sr km^{-2} y^{-1} in the year (i): (i=0 in 1950), and λ_c is the decay factor for the various removal processes (D.1.6.1.1.).

$$A_{pn} = \sum_{i=0}^{i=n} d_i e^{-\lambda_1 (n - i + 1)}, \qquad \text{(Eq.D.1.6.2.4)}$$

where A_{pn} is the accumulated ^{90}Sr in mCi km^{-2} in the ploughing layer n years after the deposition.

$$A_{un} = A_{cn} - A_{pn} \qquad \text{(Eq.D.1.6.2.5)}$$

where A_{un} is the accumulated ^{90}Sr in mCi km^{-2} below the ploughing layer

$$A_{un} = \sum_{i=1}^{i=n} d_i^{*} e^{-\lambda_r (n - i + 1)} \qquad \text{(Eq.D.1.6.2.6)}$$

where λ_r is the radioactive decay factor (for ^{90}Sr: 0.025 y^{-1}) and

$$d_i^{*} = (1 - e^{-\lambda_p}) \cdot A_{p(i-1)} \qquad \text{(Eq.D.1.6.2.7)}$$

where d_i^{*} is the transfer rate of ^{90}Sr in mCi km^{-2} y^{-1} from the ploughing layer to the lower-lying soil layers; d_i^{*} may thus be considered as a "secondary fallout rate" to the undisturbed soil layers below the ploughing layer.

Let us assume that the relative root uptake of ^{90}Sr by crops is proportional to the accumulated ^{90}Sr, and that it furthermore depends upon the median depth according to Eq.D.1.6.2.1. If the cultivated soil is considered as two compartments, the ploughing layer (p) and the underlaying layer (u), we may estimate the relative root uptakes (U_p) and (U_u) from each of these layers

from Eq.D1.1.6.2.1. and thereafter weight the U-values with $\frac{A_p}{A_c}$ and $\frac{A_u}{A_c}$, respectively, (cf.Eqs.D.1.6.2.4. - D.1.6.2.6.). The total relative root uptake then becomes: $U_t = \frac{1}{A_c} (A_p \cdot U_p + A_u \cdot U_u)$. The median values used for the calculation of U_p and U_u were, respectively, 10 cm and $(a'_{m_n} \sqrt{t'} + 20)$ cm. In the last quality a'_{m_n} was the secondary fallout-weighted median depth calculated analogously to a_{m_n} (cf.Eq.D.1.6.2.2.), applying Eq.D.1.6.2.7. for the calculation of the secondary fallout rate to the underlaying layer.

Table D.1.6.2.2. Calculated effective half-lives for root uptake of ^{90}Sr from cultivated soils.

Period	(units: years)		
	a = 2		
	c = 0.1	c = 0.2	c = 0.4
1950-1964	18	16	13
1965-1975	18		13
1976-1989	16	14	12
1990-2000	16		13

Cf. the remarks to Table D.1.6.2.1.

The relative root uptakes U_t for each year were then compared by a correlation analysis with the corresponding ^{90}Sr soil levels for various effective half-lives of ^{90}Sr. For c-values varying from 0.4 to 0.1, the best fits were obtained for effective half-lives of 13-18 years (table D.1.6.2.2.). In the next 25 years the effective half-life in cultivated soil will, accordingly to the model, decrease to 12-16 years, provided that the fallout rate continues to decrease. The estimated half-lives for ^{90}Sr in cultivated soil were similar to those for uncultivated soil. The decreasing tendency of the half-lives estimated in cultivated soil was due to the modifying effect of the ploughing layer, which as long as the bulk of the activity remained in this layer enhanced the relative root uptake due to the constant median depth of this layer as

compared with the decreasing median depth of untouched soil. When the fallout rate decreased, the relatively rapid downward transport of the activity from the ploughing layer reduced the effective half-life in cultivated soils.

Table D.1.6.2.3. Calcium and stable strontium in the soil of the ploughing layer collected from 10 Danish experimental farms in 1966

Location	g Ca kg^{-1}	mg Sr kg^{-1}	mg Sr (g Ca)$^{-1}$
Tylstrup	0.98	2.46	2.51
Ødum	2.62	7.39	2.82
Studsgård	1.30	4.29	3.30
Askov	1.90	6.57	3.46
Jyndevad	1.33	4.18	3.14
Blangstedgård	4.16	12.48	3.00
Tystofte	2.56	6.43	2.51
Virumgård	1.97	6.82	3.46
Abed	7.93	18.64	2.35
Åkirkeby	1.98	4.36	2.20
Mean	2.67	7.36	2.88
SE	0.65	1.52	0.15
Median	2.0	6.5	2.9

The determinations were performed on dried soil from a HCl extract.

D.2.2.1. Influence of variety on radionuclide concentrations in cereal grain

Several authors have examined the influence of species and variety on the indirect contamination of cereal crops. LEE et al. (Le65) thus observed that seeds of Thatcher wheat contained 70-75% of the ^{85}Sr concentrations found in Moncalm barley and Prolific rye. In a later study (Le69) it was observed that Thatcher wheat showed $^{85}Sr/^{45}Cs$ ratios which were only 80% of those in Chinook wheat. SMITH et al. (Sm63) found that $^{89}Sr/Ca$ ratios between 20 varieties of barley varied a factor of 2.1 and between 20 varieties of wheat a factor of 2.0.

In the present material, both the ^{90}Sr and the ^{137}Cs concentrations in winter and spring varieties did not as a whole differ significantly from each other. In the case of ^{90}Sr, the winter varieties (Starke, Cato, and Cranich) of wheat probably ($p \leq 0.05$) contained more activity than the spring varieties (Koga, Sappo, and Kolibri), which may be due to a greater root uptake of ^{90}Sr by the winter wheat (cf.fig.D.2.2.1.). A study of 17 varieties of barley collected in 1965 at Askov showed that the variance between varieties was not significantly greater than the analytical error. The lack of intervarietal difference may be due to the dominating influence of direct contamination in 1965. Considering the period 1967-1975, when the direct contamination was less important than the indirect,

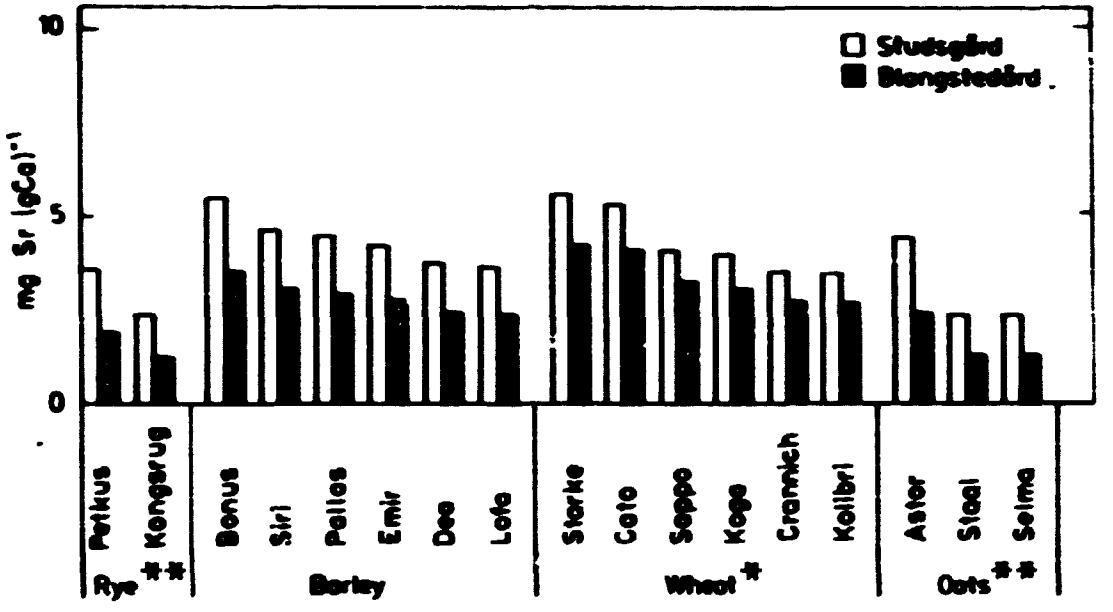

Fig. D.2.2.1. Stable Sr in varieties of Danish cereal grain collected at Studsgård and Blangstedgård (Fig. A.1.1.3.1.) during 1964-1975. The stars show the degree of significance among varieties within the various species. Studsgård showed significantly higher mg Sr (g Ca)$^{-1}$ levels than Blangstedgård.

an anova of pCi ^{90}Sr (g Ca)$^{-1}$ in wheat varieties showed that Cato wheat contained 1.6 times higher levels than Kolibri wheat. A similar anova of 7 barley varieties did not reveal any significant differences.

Anovas of the stable Sr to Ca ratio in the various varieties of the 4 grain species studied showed (fig.D.2.2.1.) that wheat and barley varieties contained the highest mg Sr (g Ca)$^{-1}$ ratios. The variation of the Sr/Ca ratio within a species varied between 1.5 and 1.9, which was close to the range observed by SMITH et al. (Sm63). If the time variation of the varieties used was studied, it appears that, for rye, Petkus has in recent years replaced Kongsrug, which was more frequent in the first years of observations; this has resulted in a slower decay in the pCi ^{90}Sr (g Ca)$^{-1}$ levels in rye than would have been expected if there had been no change in varieties. In the case of barley, the time trend has been from Bonus over Pallas to Emir and Siri. Figure D.2.2.1. indicates that this change in varieties might also have slowed down the decay rate of ^{90}Sr, although less evidently than for rye. The time trend for wheat showed that Koga and Starke have been increasingly replaced by Cranich, Cato, Kolibri, and Sappo; however, this can hardly have influenced the time trend of the ^{90}Sr/Ca ratios in wheat samples. Finally, for oats, the variety Stål was replaced by Astor, which in turn was to some degree replaced by Selma. This could have increased the ^{90}Sr/Ca ratio in oats in 1970-1972, but it may have enhanced the general decrease in the ^{90}Sr concentrations in recent years.

D.2.2.2.1. Comparison between prediction models for pCi ^{90}Sr (g Ca)$^{-1}$ in cereal grain. In earlier publications (Aa66c, X, Aa72a) various prediction models for the ^{90}Sr and ^{137}Cs concentrations in cereal grain were suggested. A comparison between the observed and the predicted pCi ^{90}Sr (g Ca)$^{-1}$ values was made in table D.2.2.2.1. using the four models proposed throughout the years. Two years were tested: 1963 when the fallout rate showed a maximum and the accumulated fallout was moderate, and 1973 when the fallout rate was low and the accumulated fallout was relatively high. The comparison shows that the predictions have generally been improved throughout the years, and that the latest models gave the best fit to the observed values in 1963 as well as in 1973. It furthermore appeared that the prediction for 1963, when the ^{90}Sr concentrations in grain were high, fitted the observations better

Table D.2.2.2.1. A comparison between observed and predicted ^{90}Sr concentrations in grain using previous and present models. The figures are the ratios of the pCi ^{90}Sr $(g\ Ca)^{-1}$ levels.

Model	Year of sampling	Rye	Barley	Wheat	Oats	Mean of species	CV
1966	1963	0.78	0.82	0.63	1.05	0.82	0.21
(Aa 66c)	1973	0.81	2.74	1.38	1.32	1.56	0.53
1968	1963	1.08	1.04	0.92	1.00	1.01	0.07
(X)	1973	0.89	1.48	1.33	1.02	1.18	0.23
1972	1963	1.15	0.92	0.98	1.06	1.03	0.10
(Aa 72a)	1973	0.56	0.83	0.91	0.61	0.73	0.23
1974	1963	1.02	0.99	1.03	1.00	1.01	0.02
(RRD 75)	1973	1.27	1.05	0.99	1.00	1.08	0.12
Mean of previous models	1963	1.01 CV:0.16	0.94 CV:0.10	0.89 CV:0.20	1.03 CV:0.03	0.97 CV:0.10	
	1973	0.88 CV:0.33	1.53 CV:0.56	1.15 CV:0.21	0.99 CV:0.29	1.14 CV:0.30	
Present models (cf. Table C.2.2.3 Nos. 8, 10, 12, 14)	1963	1.02	0.99	1.03	1.00	1.01	0.02
	1973	1.28	1.04	0.99	1.01	1.08	0.12

than the 1973 predictions when the levels were low. This was partly due to the fact that 1973 was incorporated in the last of the models only, whereas 1963 was represented in all models. Furthermore, the analytical error of the 1973 figures (due to the low activity levels) was greater than that of the 1963 determinations (the residual error was thus 20% in 1973, but 11% in 1963).

D.2.2.2.2. Error estimate of prediction models for grain. To assess the uncertainty of the radioecological sensitivities estimated from the prediction models, the material was divided into even and odd years. Thereafter the prediction models and the corresponding sensitivities for each set of years were calculated. These calculations showed in the case of grain that the relative standard error of the mean of the two determinations was approx. 10%. A similar error was found for the mean sensitivity determined by the different models applied to the same sample type (cf.tables C.2.2.1. - C.2.2.6.).

D.2.4.1. The calcium and potassium content of Danish, Faroese and Greenlandic grass samples

As for other vegetable products in this study, the radionuclide content of grass referred to fresh weight. Furthermore, for ^{90}Sr and ^{137}Cs, the radionuclide content referred to the contents of Ca and K, respectively. Contrary to most other vegetable samples, the concentrations of Ca and K varied in the grass samples received, partly due to a varying water content of the samples and partly due to environmental factors. Where the Danish grass generally consisted of fresh green vegetation, the Faroese and especially the Greenlandic samples were generally withered hay.

Table D.2.4.1. Calcium and potassium in Danish, Faroese and Greenlandic grass samples (as received).

Area	g Ca kg^{-1} ± 1 SD (determinations and period)	g K kg^{-1} ± 1 SD (determinations and period)	References
Denmark	1.24±0.34 (19; 1963)	7.0±3.2 (55; 1964-70)	(RRD 63, RRD 59-76)
Faroes	1.5±1.1 (21; 1964-73)	8.4±6.2 (20; 1964-72)	(RRF 62-76)
Greenland	3.3±1.9 (34; 1963-73)	12±8.8 (30; 1963-73	(RRG 62-76)

The dry matter content of Danish grass was estimated at 20% from the Ca content in this table and from the mean Ca content (g kg^{-1} dry matter) in rye-grass, cock's foot, timothy and meadow fescue determined by ANDERSEN (An 67a). The mean yield in kg fresh weight per m^2 was determined in June and September 1963 at the 10 state experimental farms in Denmark as 0.74±0.24 kg m^{-2} (1 SD).

The calcium concentrations in fresh grass were assumed to be equal to those in Danish grass, i.e. 1.24 g Ca kg^{-1} (Table D.2.4.1.), while the potassium concentrations differed among the three regions. In the fresh Faroese grass the potassium content was estimated as: $\frac{8.4 \cdot 1.24}{1.5} = 6.94$ g K kg^{-1} and in the Greenlandic samples as: $\frac{12 \cdot 1.24}{3.3} = 4.51$ g K kg^{-1}.

These values were used in the calculation of the radioecological sensitivities given in the text (2.4.1.).

Table D.3.2.1. Feed units of various crops relative to those from grass.

Area of country (zone) (cf. Fig. A.1.4.2.1.)	Fodder beets / grass	Factory refuse / grass	Beet tops / grass	Straw / grass	Feed units from grass **) / number of cows *)
I	0.54	0	0.11	0.24	3000
II	0.60	0	0.12	0.30	2900
III	0.51	0	0.11	0.21	4200
IV	0.37	0.01	0.08	0.23	4100
V	0.39	0.09	0.18	0.32	2700
VI	0.42	0.09	0.16	0.41	3200
VII	0.22	0.90	0.60	0.52	2700
VIII	0.52	0.01	0.13	0.31	2900

The ratios were the mean of the 1966 and 1971 production data (Da 57-77). Factory refuse mostly consists of beet-offal from sugar production and of molasses. The feed unit of factory refuse was estimated at 1/3 of the feed unit of sugar beet for sugar production (Da 57-77).

*) cows and heifers having calved

**) total grass production

D.3.4.4. On the fallout rate used in the prediction models for samples from West Greenland

In the prediction models for samples from the west coast of Greenland, e.g. lichen, grass, sheep and reindeer, the fallout rates measured at Godthåb were used as the only available relevant data. However, the amounts of precipitation decrease by nearly a factor of four along the west coast from Julianehåb - Godthåb in the south to Egedesminde in the north (Da63-76). The fallout rate may thus have been overestimated in the area of Greenland from where the samples were obtained, hence the radioecological sensitivity of the samples may have been underrated. On the other hand, no local variation was observed in the lichen and grass samples or in the reindeer. This suggested that dry fallout compensated for the lack of wet deposition at the northern locations (cf.1.3.1.), and that the fallout rate measured at Godthåb may thus be a reasonable estimate of the total deposition on the west coast of Greenland.

D.3.5.1. Calcium content of cod-flesh

Before the ^{90}Sr determinations of fish flesh, the bones were removed as far as possible. However, for the samples received as whole fish the bone content of the flesh used for analysis was generally higher than for samples received as fillets. The samples of cod from Denmark and Greenland were whole fish and the flesh contained 0.90 ± 0.26 g Ca kg^{-1} (1 SD, 28 samples); the Faroese codfish samples, on the other hand, were fillets with a concentration of 0.11 ± 0.03 g Ca kg^{-1} (79 samples) only. As the ^{90}Sr content of the flesh was proportional to the calcium concentration, because the $^{90}Sr/Ca$ quotients in bone and flesh were similar, it was most expedient to apply pCi ^{90}Sr $(g\ Ca)^{-1}$ when the ^{90}Sr concentrations in codfish from the various areas were compared.

D.4.2.1. The importance of the variations in the composition of the Danish total diet.

General comments to tables D.4.2.1.a. and D.4.2.1.b.

The "1962 diet composition" was identical to the "3000 kcal diet" proposed at that time by HOFF JØRGENSEN (RRD62). The 1975 composition was the annual per caput consumption in Denmark in 1975 according to Danmarks Statistik (Da57-77) (cf., however, the special remarks). The joule contents of the various diet constituents was estimated from (Da68). The Ca, Sr and K contents were the mean values determined throughout the years in the Risø diet studies (RRD59-76), except for the stable Sr contents in milk products, meat and eggs, which were estimated from UK data (Ag61) (cf. also 4.2.3.). The ^{90}Sr and ^{137}Cs levels were those measured in 1976.

Table D.4.2.1.a. The contents of radionuclides and stable elements in the Danish total diet of 1962 composition collected in 1976.

Type of food	Annual per caput quantities according to 1962 diet composition						
	kg	GJ	g Ca	mg Sr	g K	1976 pCi ^{90}Sr	1976 pCi ^{137}Cs
Milk	164	0.46	197	59	262	672	705
Cheese	9.1	0.12	75	23	11	263	30
Grain products[a]							
rye flour	29.2	0.40	119	242	134	599	458
wheat flour	43.8	0.60	117	260	88	169	177
grits (oat)	7.3	0.12	24	31	14	88	81
Potatoes	73	0.27	4	12	270	124	161
Vegetables[f]	43.8	0.04	18	58	92	289	123
Fruit	51.1	0.11	4	12	72	118	102
Meat	54.7	0.57	6	4	175	22	1384
Eggs	10.9	0.07	6	8	8	10	29
Fish[b]	10.9	0.03	8	28	36	5	643
Coffee and tea[c]	5.5	-	3	15	94	40	196
Beer, wine, etc.[d]	75	0.11	8	21	25	11	40
Water[e]	478	-	34	310	-		-
Sugar	54.8	0.90	-	-	-	-	-
Fats, butter, etc.	29.2	0.90	-	-	-	-	-
Total		4.70	623	1083	1281	2410	4129
Daily mean			1.71	2.97	3.51	6.6	11.3
				mg Sr (g Ca)$^{-1}$ = 1.74		pCi ^{90}Sr (g Ca)$^{-1}$ = 3.9	pCi ^{137}Cs (g K)$^{-1}$ = 3.2

Special remarks to tables D.4.2.1.a. and D.4.2.1.b.

a) The Ca figures for grain products included creta praeparata, i.e. 107 g Ca y^{-1} in rye flour, 108 g Ca y^{-1} in wheat flour and 20.6 g Ca y^{-1} in grits. The stable Sr contents in the creta preparata were 209, 232 and 25 mg y^{-1}, respectively. The creta praeparata used in rye and wheat flour was calcium carbonate, while dicalcium phosphate was used in oat grits (Da68).

b) The fish consumption given for the 1975 diet corresponded, as in the case of the 1962 diet, to the net consumption of fish. The figure in the 1975 diet was thus approx. 5 kg y^{-1} less than the figure in Danmarks Statistik (Da57-77), which was based on the amount of whole fish.

Table D.4.2.1.b. The contents of radionuclides and stable elements in the Danish total diet of 1975 composition collected in 1976.

Type of food	Annual per caput quantities according to 1975 diet composition						
	kg	GJ	g Ca	mg Sr	g K	1976 pCi ^{90}Sr	1976 pCi ^{137}Cs
Milk	175	0.49	210	63	280	718	753
Cheese	9.6	0.13	79	24	12	277	30
Grain products[a]:							
rye flour	20.4	0.28	83	168	94	418	320
wheat flour	40.5	0.56	108	240	81	156	164
grits (oat)	7.2	0.12	23	30	14	87	80
Potatoes	75	0.28	4	12	278	128	165
Vegetables[f]	49.4	0.05	18	58	104	326	138
Fruit	51.7	0.11	4	12	72	119	103
Meat	69.3	0.72	7	4	222	28	1753
Eggs	11.4	0.08	6	9	8	10	31
Fish[b]	15	0.05	11	39	50	8	885
Coffee and tea[c]	8.3	-	5	25	156	57	313
Beer, wine, etc.[d]	110	0.18	11	31	36	11	58
Water[e]	438	-	28	248	-		-
Sugar	47.5	0.78	-	-	-	-	-
Fats, butter, etc.	29.2	0.90	-	-	-	-	-
Total		4.73	597	963	1407	2343	4793
Daily mean			1.64	2.64	3.85	6.4	13.1
				mg Sr (g Ca)$^{-1}$ = 1.61		pCi ^{90}Sr (g Ca)$^{-1}$ = 3.9	pCi ^{137}Cs (g K)$^{-1}$ = 3.4

c) The coffee grounds and the used tea leaves contained some of the Sr and Ca present in the water used for the extraction. An experimental tracer study with ^{85}Sr and ^{47}Ca showed that coffee grounds absorbed 77% of the Ca and 75% of the Sr in the water. In the case of tea, the leaves absorbed 22% Ca and 17% Sr. It was further observed that 0.68 g Ca and 3 mg Sr per kg coffee beans were extracted into the liquid coffee. For tea, the corresponding figures were 0 g Ca and 2 mg Sr kg^{-1} leaves. In 1962 the annual mean consumptions (RRD62) were 3.67 kg coffee and 1.83 kg tea per caput. In 1975 the consumptions were estimated at 8 kg coffee and 0.3 kg tea per caput (Da68).

d) Samples of beer (Faxe, Carlsberg) and red wine (Spanish) showed mean levels of 0.10 g Ca l^{-1} and 0.28 mg Sr l^{-1}. As the levels in beer and wine were similar, and as beer was more important in the Danish diet than wine, only beer is considered in the table. In a Japanese study of ^{137}Cs in beer the potassium

content was measured at 0.33 g K l^{-1} (Sh68). This figure was used and it was assumed that the $^{137}Cs/K$ ratio in Danish beer from a given year equalled that in Danish barley of the preceding year.

e) The annual per caput intake of water inclusive beer, wine, etc., was estimated at 548 l (1.5 l day^{-1}). The mean concentrations in Danish drinking water were 0.09 g Ca l^{-1} and 0.79 mg Sr l^{-1}. Assuming that 20 l water were required to make coffee from 1 kg coffee beans, and 100 l to make tea from 1 kg tea leaves (Da68), the Ca and Sr figures in the table could be calculated from the experimental data in c). If the coffee and tea were prepared as in the total diet analysis (where all water included in the analysis was used for the extraction), a tracer experiment showed that the Ca and Sr levels of the water included in the total diet sample would have been 63% of the original water content, i.e., in the 1962 diet: 27 g Ca and 238 mg Sr per annum. Taking this into account, the expected daily intakes according to the total diet measurements became 1.69 g Ca and 2.77 mg Sr, and the mg Sr (g Ca)$^{-1}$ ratio was 1.64. In the direct measurements on total diet (1961-1976), means of 1.73 g Ca d^{-1} and 1.52 mg Sr (g Ca)$^{-1}$, or 2.63 mg Sr d^{-1} (cf. 4.2.3), were observed. The direct measurements were thus compatible with the calculated values, and the estimates of the absorption of Ca and Sr from the drinking water in coffee-grounds and tea-leaves were thus confirmed.

f) The daily mean consumption of vegetables consisted of 50 g leaf vegetables, 30 g root vegetables and 40 g peas and beans (RRD62). It was assumed that the relative proportions between the three vegetable groups were unchanged in the 1975 diet composition.

D.4.2.2. Assessment of the radioactive contamination of the Danish total diet

The annual mean of year (i) for the total diet ("Diet C") was obtained as the weighted mean of the samplings in December (i-1), June (i) and December (i). The December samplings were each weighted by one, while the June samplings carried a weight

of two. Using this method all samples had the same weight in the material as a whole, because the December samples always contributed to two years.

The assessments of ^{90}Sr and ^{137}Cs in the Danish total diet have, in general, given the same results both for "Diet C" and for "Diet P" (cf.fig.4.2.2.). The correlation coefficients between the methods were for ^{90}Sr: 0.993 (15 years of observations) and for ^{137}Cs: 0.986 (14 years of observations), i.e. both highly significant. The mean ratios between the annual country means obtained from the direct measurement of a mixed total diet sample ("Diet C") and from measurements of the single components ("Diet P") were 1.05 $\pm$ 0.12 (1 SD) for pCi ^{90}Sr (g Ca)$^{-1}$ and 1.00 $\pm$ 0.19 for pCi ^{137}Cs (g K)$^{-1}$.

Approximately 25% of the Danish population live in the Copenhagen area where food production is negligible. The food consumed by the Copenhagen population may originate from the entire country. It was thus not obvious that the radionuclide concentrations of the Copenhagen diet should correspond to those in the Zealand diet (zone VI). The mean ratio during 1962-1976 between ^{90}Sr in the Copenhagen diet and in the diet from the remainder of Zealand was 1.05 $\pm$ 0.21 (1 SD) (df = 29) (pCi ^{90}Sr (g Ca)$^{-1}$ figures), and for pCi ^{137}Cs (g K)$^{-1}$ the ratio was 1.22 $\pm$ 0.44 (df = 27). The ^{90}Sr diet levels were not significantly different between Copenhagen and Zealand, but the ^{137}Cs concentration was probably higher in the Copenhagen diet, suggesting a transfer of foods from Jutland to Copenhagen.

An unweighted mean of the radionuclide concentrations in locally produced foods from the 8 zones (Table A.1.4.2.) may underestimate the average diet intake by the Danish population, because the four zones in Jutland, where the activity levels were higher, produced more than half of the diet consumed in Denmark. In the case of a total diet that also includes non-locally grown diet components (e.g. bread), the unweighted zone mean may also be lower than the population weighted mean, especially as a result of the transfer of foods from Jutland to Copenhagen. However, a comparison between the population

weighted means of ^{90}Sr and ^{137}Cs in the Danish diet and the unweighted means of the levels in the 8 zones did not show any significant difference.

It was thus concluded that whether use was made of population weighted means or the unweighted zone means of the direct total diet measurements ("Diet C"), or whether the concentrations obtained from single diet constituents were applied ("Diet P"), the estimates of the ^{90}Sr and ^{137}Cs in the total diet were not significantly influenced.

Table D.4.2.2. A comparison of the relative contributions from the various diet groups and of the transfer coefficients P_{23} (I.e. the radioecological sensitivity) from deposition to the total Danish diet of ^{90}Sr and ^{137}Cs assessed by UNSCEAR (Un 77) and by the present study (Risø).

	Strontium-90		Cesium-137	
	Risø	UNSCEAR	Risø	UNSCEAR
Milk products	30%	29%	17%	19%
Cereal products	42%	44%	39%	38%
Vegetables, potatoes and fruits	26%	25%	10%	10%
Meat, fish and eggs	2%	2%	34%	33%
P_{23} calculated from the measurements of the individual diet groups (cf. 4.2.2 and Table 4.2.2)	4.78	4.70	4.36	4.58
P_{23} assessed from the total diet (cf. 4.2.2 and Tables C.4.2.1 and C.4.2.2)	5.1 ± 0.33	4.01	4.4 ± 0.15	4.08
Units for P_{23}	pCi ^{90}Sr (g Ca)$^{-1}$ · y per mCi ^{90}Sr km^{-2}		pCi ^{137}Cs (g K)$^{-1}$ · y per mCi ^{137}Cs km^{-2}	

D.4.3.2. Prediction models based on diet levels for ^{90}Sr in adult human bone

Instead of the prediction models presented in figs.4.3.2.1. and 4.3.2.2, one may calculate two models for adult bone $\geq$ 30 years omitting the requirement that all the individuals should be born before 1935. The equations for pCi ^{90}Sr (g Ca)$^{-1}$ in adult human vertebrae from Jutland and the Islands, respectively, then became:

$$\text{pCi } {}^{90}\text{Sr (g Ca)}^{-1}_{(i)} = 0.019 \text{ pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{(i)} + 0.028 \sum_{m=1}^{m=\infty} \text{pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{i-m} \cdot e^{-0.23m}$$

(Eq.D.4.3.2.1)

$$\text{pCi } {}^{90}\text{Sr (g Ca)}^{-1} = 0.023 \text{ pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{(i)} + 0.023 \sum_{m=1}^{m=\infty} \text{pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{i-m} \cdot e^{-0.173m}.$$

(Eq.D.4.3.2.2)

The transfer factors estimated from these equations were: 0.14 and 0.15 pCi ^{90}Sr (g Ca)$^{-1}$ · y in bone per pCi ^{90}Sr (g Ca)$^{-1}$ y in diet, i.e. nearly the same as those observed in figs. 4.3.2.1. - 4.3.2.2., but the correlation between observed and calculated values was a little poorer than those in figs. 4.3.2.1. - 4.3.2.2. (Jutland r = 0.9315 against r = 0.9343 and the Islands r = 0.8249 against r = 0.8835).

If the age group 20-29 y was considered, the result for the entire country was:

$$\text{pCi } {}^{90}\text{Sr (g Ca)}^{-1}_{i} = 0.036 \text{ pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{(i)} + 0.056 \sum_{m=1}^{m=\infty} \text{pCi } {}^{90}\text{Sr (g Ca)}^{-1} \text{ in Diet}_{i-m} \, e^{-0.41m} \quad \text{(Eq.D.4.3.2.3)}$$

The transfer factor for ^{90}Sr from diet to bone for this age group became P_{34} = 0.17 pCi ^{90}Sr (g Ca)$^{-1}$ y in bone per pCi ^{90}Sr (g Ca)$^{-1}$ y in the diet, and the correlation coefficient between observed and calculated values was r = 0.9454. The turnover rate of the 20-29 y age group was 0.31 y^{-1} (cf.Eq. 4.3.3.) corresponding to a biological half-life of 2.2 y.

D.4.4.1. Influence of the time of tooth sampling on the ^{90}Sr concentrations observed in shed deciduous teeth

To determine whether the ^{90}Sr concentrations in shed deciduous teeth from a given birth year were influenced by the time of the sampling, anovas were carried out on the various types of tooth collected in the four sampling periods: a) 1962-1965, b) 1966-1969, c) 1970-1973 and d) 1974-1976. In the case of incisors and cuspids, it appeared that teeth from a given birth year showed significantly higher levels when they were from period c) than from b) and d), and further that period a) showed lower levels than b) (cf. also XII). First and second molars did not show any significant difference among the various sampling periods. In the Faroese and Greenlandic material the tendency was the same. The difference observed between sampling periods was explained by the fact that teeth from a given birth year, received in the preceding period, mostly originated from children born in the first part of this birth year, while teeth from a subsequent period generally originated from children born in the last part of the birth year. As the ^{90}Sr concentrations generally increased until 1963, also within the years, and then decreased, the tendency observed among sampling periods becomes intelligible. The shed deciduous teeth have thus not shown any significant tendency to an exchange of the tooth ^{90}Sr with ^{90}Sr introduced into the environment after the formation of the tooth.

D.4.7.7. Collective effective dose equivalent commitment

The International Commission on Radiological Protection (ICRP) (Ic77) has defined the quantity effective dose equivalent, H_E, as

$$H_E = \sum_T w_T H_T , \qquad \text{(Eq. D.4.7.7.1)}$$

where H_T is the dose equivalent in tissue T and

w_T is the weighting factor for each tissue and represents the ratio of the stochastic risk from irradiation of tissue, T, to that for the whole body when uniformly irradiated.

In the derivation of w_T the ICRP chose to consider the risk of fatal cancer in all body organs and tissues (other than in skin) and the hereditary effects in the first two generations.

The collective effective dose equivalent, S_E, is given by

$$S_E = \int_0^\infty H_E \, N(H_E) dH_E \qquad \text{(Eq. D.4.7.7.2)}$$

where $N(H_E)dH_E$ is the number of individuals receiving an effective dose equivalent in the range H_E to H_E+dH_E

Collective effective dose equivalent commitment, S_E^C, is given by:

$$S_E^C = \int_0^\infty \dot{S}_E(t) dt \qquad \text{(Eq. D.4.7.7.3)}$$

where $\dot{S}_E$ is the collective dose equivalent rate. This quantity is taken as a measure of the total health detriment from exposure of a population to ionizing radiation (cf. the above qualifications).

D. 4.7.8. The significance of the time distribution of fallout when applied for calculation of the radioecological sensitivity

The infinite time integral, IT, of the radionuclide concentration in a sample from a given contamination of the environment depends upon the seasonal distribution of the contamination. This is especially evident for shortlived radionuclides. A contamination of the pastures with ^{131}I during winter will thus hardly influence the contamination of milk in the Nordic countries because the cows are eating stored fodder at that time of the year. For certain nuclides in certain crops, e.g., for ^{90}Sr in cereal grain, IT will be significantly higher if the contamination occurs just before harvest than at any other time of the year (cf. 2.2).

The radioecological sensitivity, RS, is defined as the infinite time integral in the sample considered arising from a unit deposition of 1 mCi km^{-2} - of the radionuclide considered - distributed throughout the year as has been the case for ^{90}Sr on the average in the period 1962-1974, i.e. in January-February: 8%, in March-April: 17%, in May-June: 32%, in July-August: 25%, in September-October: 10%, and in November-December: 8%. In the case of shortlived radionuclides, e.g., ^{89}Sr and ^{131}I, for which the seasonal distribution have been highly variable (because these radionuclides are strongly influenced by the fresh tropospheric deposition), we have calculated the radioecological sensitivity assuming a uniform distribution throughout the year.

Let us assume that we have uniform distributions of ^{137}Cs and ^{90}Sr throughout the year, which with approximation may be the case if we deal with operational releases from nuclear plants. For most samples the IT from an annual unit deposition would then be less than the RS, because the global fallout (used for the calculation of RS) during November-April is half of that during the summer season, when the production of crops takes place. From the prediction models we estimate that in case of ^{137}Cs: IT ~ 0.7 RS and for ^{90}Sr: IT ~ 0.85 RS. In other words, if radioecological sensitivity is applied to releases uniformly distributed throughout the year the infinite time integrals of such releases are overestimated.

In case of short-time deposition of longlived substances, e.g. by accidental releases, the seasonal influence on the consequences is significant, especially for radionuclides with a low root uptake such as ^{137}Cs. Deposition of this radionuclide during the winter season will in Denmark have only little influence on dietary ^{137}Cs.

D. 5.2.1. Statistical information on Danish agricultural produce with estimates of calcium and potassium levels

The Danish Statistical Office (Da57-77) publishes annual statistics on agriculture, gardening and forestry. The calculations of the inventories and transfers within the Danish agricultural

ecosystem of ^{90}Sr and ^{137}Cs (cf. 5.2) were based on the statistical data from 1975 assuming they represented a steady state situation. In the following tables the figures for production and use of Danish agricultural produce is shown, for animal produce together with the Ca and K levels in the various products. For the calculation of the inventories and transfers of ^{90}Sr and ^{137}Cs the data in these tables were used together with the pertinent radioecological sensitivities shown in Appendix C.

Table D.5.2.1 deals with the vegetable agricultural produce. It appears that the Danish agriculture is nearly self-supporting with vegetable produce. The import of concentrates is approximately cancelled out by the export of grain and various seeds. In the main text we have assumed that the import and export of ^{90}Sr and ^{137}Cs with vegetable produce counterbalance each other, because an estimation of ^{90}Sr and ^{137}Cs in imported fodder is doubtful. The remainder part of the table indicates various losses (e.g. strawburning in the field), use for industrial purposes (e.g. building materials) and use for food production (e.g. sugar from beets). The various amounts in the table do not necessarily balance each other exactly because the use may be both greater or lesser than the production and import together for the year 1975. However, the deviations are immaterial in the present context.

Danish animal agricultural produce is dealt with in Table D.5.2.2. The calcium concentration in the various animals is 1.7% of the living weight and the potassium concentration is 0.35% (We79). Beef and pork contain 0.1 g Ca kg^{-1} and 3.2 g K kg^{-1} (RRD59-76), bone and meat meal contains approx 104 g Ca kg^{-1} (Br69) and marrow-free bone contains 150 g Ca kg^{-1} (Ic59).

The annual milkproduction in Denmark and its use are shown in Table D.5.2.3 together with the Ca and K levels in the various products. In the table imports have been neglected because it is immaterial (the import of cheese, is for example, approx 3% of the export). Total milk represents the total production of milk; hence the sums of the Ca and K contents of the various milk products should be equal to the calculated Ca and K levels of the total milk. This is, as the table shows, the case to a good approximation.

Table D.5.2.1. Production and use of Danish vegetable agricultural produce

	Production		Fodder	Export	Import	Danish human consumption	Remainder
	Area km^{-2}	Amount Tg	Tg	Tg	Tg	Tg	Tg
Rye grain	500	0.163	0.035	0.021	-	0.103[b]	
Wheat grain	1020	0.52	0.085	0.23	0.015	0.205[b]	
Barley grain	14600	5.2	4.6	0.75	0.027	-	
Oats grain	1110	0.37	0.38	0.007	0.008	0.013[b]	
Total grain	17200	6.3	5.3[g]	1.0	0.05	-	
Straw	17200	5.3	1.64	-	-	-	3.7
Grass etc.	9000	23[i]	20	0.13[h]	-	-	2.9
Beet tops	3000	4.0	3.4	-	-	-	0.6
Roots	3000	12.4	10[f]	0.042[e]	-	0.38[e]	2
Imported concentrates[a]	-	-	1.4	-	1.4[c]	-	-
Vegetables	67	0.185	-	0.014	0.071	0.25	-
Fruits	100	0.157	-	- [d]	0.122[d]	0.27	-

[a] Includes maize and oil-cakes and -meal.

[b] Flour and meal; the bran is partly used as fodder.

[c] Estimated equal to fodder figure.

[d] Export of fruits was subtracted from the import.

[e] Potatoes.

[f] 0.32 Tg of molasses, pulp and dregs from the sugar production is used as fodder and included in the 10 Tg.

[g] 0.16 Tg bran is included in the total grain figure for fodder.

[h] rape and various seeds of grass.

[i] Assuming 1 feed unit equals 7.5 kg grass (Da57-77).

Table D.5.2.2. Production and use of Danish animal agricultural produce with estimates of calcium and potassium contents

	Production			Fodder			Human diet			Export[d]			Remainder[e]	
	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg	Ca Gg	K Gg
Cattle	0.501	8.5	1.75	-	(5.6)[b]	(0.29)[b]	0.091	0.009[c]	0.29	0.154	2.6	0.54	0.3	0.63
Pigs	0.981	16.7	3.4	-	(5.6)[b]	(0.29)[b]	0.216	0.022[c]	0.69	0.560	9.5	1.96	1.6	0.46
Poultry	0.106	1.8	0.37	-	-	-	0.039	0.003[c]	0.117	0.057	0.97	0.20	0.8	-
Eggs[f]	0.074	1.23	0.05	-	-	-	0.058	0.029[c]	0.041	0.016	0.27	0.01	0.9	0
Bone and meat meal	0.108	11.2	0.58[a]	0.108	11.2	0.58	-	-	-	-	-	-	0	0

[a] Assuming 1 kg soft tissue yielding 0.2 kg meat-meal and that 31% of the bone- and meat-meal being meat-meal (estimated from the Ca content).

[b] Calculated from bone- and meat-meal assuming that one half comes from cattle and the other half from pigs (the slaughter loss being greater for cattle than for pigs).

[c] Exclusive of bones and shells.

[d] Inclusive of bones and shells.

[e] The remainder consists mostly of waste from households (e.g. bones and shells) but includes also goods used in industry (e.g. skin).

[f] The weight of the egg shell is 10.5% of the total egg. The Ca content of the shell is 15% and the soft parts contain 0.5 g Ca kg^{-1} and 0.7 g K kg^{-1} (RRD59-76).

Table D.5.2.3. The Danish milk production and its use with estimates of the calcium and potassium contents in the various products

	Total production			Fodder			Human consumption			Export		
	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg	Tg	Ca Gg	K Gg
Whole milk and cream[a)]	0.71	0.86	1.14	0.125	0.15	0.20	0.56	0.67	0.89	0.033	0.04	0.05
Skimmed milk and butter milk[b)]	1.52	1.90	2.51	1.31	1.64	2.16	0.183	0.23	0.30	-	-	-
Dried milk[c)]	0.123	1.48	1.97	0.024	0.29	0.38	-	-	-	0.099	1.19	1.58
Whey[d)]	1.24	0.41	2.09	1.23	0.41	2.09	-	-	-	-	-	-
Cheese[e)]	0.148	1.26	0.18	-	-	-	0.049	0.42	0.06	0.099	0.84	0.12
Butter[f)]	0.139	0.02	0.03	-	-	-	0.040	0.01	0.01	0.098	0.01	0.02
Total milk[a)]	4.9	5.9	7.9	-	-	-	-	-	-	-	-	-

a) These products contained 1.2 g Ca l^{-1} and 1.6 g K l^{-1} (RRD59-76).

b) These products contained 1.25 g Ca l^{-1} and 1.65 g K l^{-1} (Da68).

c) As 1 kg dried milk ~ 10 l fresh milk, the Ca and K contents were 10 times those of the fresh milk.

d) The Ca and K levels in whey were calculated from the levels in milk and in cheese.

e) Cheese contained 8.5 g Ca kg^{-1} and 1.2 g K kg^{-1} (RRD59-76).

f) Butter contained 0.15 g Ca kg^{-1} and 0.2 g K kg^{-1} (Da68).

ABBREVIATIONS AND UNITS

J: joule: the unit of energy; 1 J = 1 Nm (= 0.239 cal)
Gy: gray: the unit of absorbed dose = 1 J kg^{-1} (= 100 rad)
Sv: sievert: the unit of dose equivalent = 1 J kg^{-1} (= 100 rem)
Bq: becquerel: the unit of radioactivity = 1 s^{-1} (= 27 pCi) *)

cal: calorie = 4.186 J
rad: 0.01 Gy
rem: 0.01 Sv
Ci: curie: $3.7 \cdot 10^{10}$ Bq (= $2.22 \cdot 10^{12}$ dpm)

T: tera:10^{12}
G: giga:10^{9}
M: mega:10^{6}
m: milli:10^{-3}
μ: mikro:10^{-6}
n: nano:10^{-9}
p: pico:10^{-12}
f: femto:10^{-15}
a: atto:10^{-18}

cap.: caput: (per individual)
TNT: trinitrotoluol; 1 Mt TNT: nuclear explosives equivalent to 10^{9} kg TNT.

cpm: counts per minut
dpm: disintegrations per minut
OR: observed ratio
CF: concentration factor
Statistical symbols: cf. appendix B.
Symbols used in prediction models: cf. appendix C.

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*) In the present study the curie unit has been applied for activity instead of the becquerel. Otherwise the comparison with other data would not have been feasible. In the conclusions, however, the activities are also given in becquerel.
Notice: mBq y $(kg)^{-1}$ per Bq m^{-2} equals pCi y $(kg)^{-1}$ per mCi km^{-2}

APPENDED REPRINTS

I Aarkrog A. (1960). Specific fallout activity in precipitation as a function of sampling height. Nature 188, 482-483

II Aarkrog A. (1964). Variation in the ratio of strontium-89 to strontium-90 in precipitation samples with sampling height. Nature 204, 771

III Aarkrog A. and Lippert J. (1967). Europium-155 in debris from nuclear weapons. Science 157, 425-427

IV Aarkrog A. (1971). Radioecological investigations of plutonium in an arctic marine environment. Health Phys. 20, 31-47

V Aarkrog A. (1977). Environmental behaviour of plutonium accidentally released at Thule, Greenland. Health Phys. 32, 271-284

VI Aarkrog A. (1969). On the direct contamination of rye, barley, wheat and oats with ^{85}Sr, ^{134}Cs, ^{54}Mn and ^{141}Ce. Radiat. Bot. 9, 357-366

VII Aarkrog A. and Lippert J. (1971). Direct contamination of barley with ^{51}Cr, ^{59}Fe, ^{58}Co, ^{65}Zn, ^{203}Hg and ^{210}Pb. Radiat. Bot. 11, 463-472

VIII Aarkrog A. (1975). Radionuclide levels in mature grain related to radiostrontium content and time of direct contamination. Health Phys. 28, 557-562

IX Aarkrog A. (1978). Variation of direct plutonium contamination in Danish cereal grains. Health Phys. 35, 489-491

X Aarkrog A. (1971). Prediction models for strontium-90 and caesium-137 levels in the human food chain. Health Phys. 20, 297-311

XI Aarkrog A. (1968). Strontium-90 in shed deciduous teeth collected in Denmark, the Faroes and Greenland from children born in 1950-1958. Health Phys. 15, 105-114

XII Aarkrog A. (1971). Prediction models for ^{90}Sr in shed deciduous teeth and infant bone. Health Phys. 21, 803-809

XIII Aarkrog A. (1963). Cesium-137 from fallout in human milk. Nature 197, 667-668

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Specific Fall-out Activity in Precipitation as a Function of Sampling Height

In 1956–57 Herbst *et al.*[1] found a definite increase with height in the fall-out activity in vegetation from an altitude of 800–1,000 m. and upwards. This was ascribed partly to the slow growth of the vegetation with increasing height and consequently greater accumulation of activity on it, and partly to greater fall-out at increased heights. Small[2] suggested in 1958 from examination of the variations in the natural activity of the air (radium B and C), compared with the meteorological conditions at ground-level, that during inversion conditions the vertical transport of bomb debris from above would decrease. By comparing the fall-out in a precipitation collector situated in the top of a 123-m. mast with that in corresponding collectors at ground-level, I found[3] for the period September 1958–March 1959 more fall-out in the former than in the latter.

It was therefore decided to examine the fall-out activity in precipitation collected in different heights. At Risö, a 123-m. meteorological mast has been built for micrometeorological measurements[4]. At heights of 0, 7, 23, 39, 56, 72, 96 and 123 m. I fixed, at a distance of 1 m. out from the mast, eight 1-litre polyethylene bottles each provided with a 154 cm.^{2} polyethylene funnel. The bottles were changed on the first of every month.

After the bottles had been collected, the amount of rain-water in each was measured and strontium-, cæsium- and cerium-carrier was added. The content of each bottle was then analysed for strontium-90 and -89, cæsium-137 and cerium-144 by the method of Osmond *et al.*[5]. The activities are given in pc./m.^{2}, and in pc./l., and are traced to the middle of the month. All the isotopes were determined for each of the ten months March–December 1959, except strontium-89 which was too weak in activity in the last months of the year for a reliable determination. A table with all the results of the measurements is given in Risö Report No. 14 (ref. 6). Table 1 gives the annual average values for the specific activity at the different heights.

A regression analysis on the figures in Table 1 shows for all the isotopes an increase in specific activity with the height. Strontium-90 displays

Table 1. AVERAGE SPECIFIC ACTIVITY IN PRECIPITATION FOR THE PERIOD MARCH–DECEMBER 1960

Isotope / Height (m.)	Strontium-90 (pc./l.)	Strontium-89 (pc./l.)	Caesium-137 (pc./l.)	Cerium-144 (pc./l.)
0	14·9	217	14·8	195
7	14·6	170	18·9	169
22	16·4	194	20·3	175
30	16·6	224	20·9	233
58	16·2	214	22·6	233
72	15·9	205	19·2	212
88	17·9	242	22·9	248
123	18·9	257	21·2	251
Mean	16·2	210	20·9	216

an increase in activity of 15 per cent per 100 m., caesium-137 of 19 per cent, cerium-144 of 27 per cent and strontium-89 of 31 per cent.

A regression analysis of the results for the ratio strontium-89/strontium-90 (see Table 2) shows an increase in that ratio with height of 13 per cent per 100 m.

The present work shows that it is possible to detect a change in the specific activity in precipitation even within a difference in height of 100 m. (The increase in total fall-out with the height is still more significant, due to increasing amount of precipitation, when ascending[4].)

The reason for this difference in specific activity must be due to the varying contribution of dry fall-out in the different rain-collectors, as it would be difficult to imagine how the rain could be less radioactive when falling down through the air.

(Regression analysis has shown that all the results are significant at the 1–2 per cent level except caesium-137 and the ratio strontium-89/strontium-90, which are significant only at the 10 per cent level.)

At an altitude of 60 m. above the Risø area, there are on the average inversion conditions during 25 per

Table 2. AVERAGE OF RATIO OF STRONTIUM-89 TO STRONTIUM-90 FOR THE PERIOD MARCH–AUGUST 1960

Height (m.)	Ratio of strontium-89 to strontium-90
0	7·6
7	7·1
22	7·6
30	6·1
58	7·9
72	7·4
88	8·2
123	8·3

cent of the time[6]. In the lower regions inversion is more frequent, in the upper less. In accordance with Small's results[2], it would be reasonable to suppose that these inversion conditions are one of the reasons why more dry fall-out is found in the bottles placed at the upper part of the mast than in those at the lower. Another factor contributing to the difference in activity will be the increasing wind velocity with increasing height[4]; this will give rise to a greater deposit of airborne particles in the higher bottles than in the lower ones[7]. Hence we get a higher specific activity in rain when the sample is taken at an altitude of, say, 100 m. than at ground-level.

Furthermore, it seems probable that short-lived fission products from fall-out would show a more pronounced variation with height than long-lived products. This is to be expected if one assumes that the dry deposit is of greater particle-size than the deposit from rain. Larger and heavier particles would be deposited more readily than fine particles[7], and consequently the fine particles will consist preferentially of older fission products than coarse ones. As a consequence of this, the age of the fall-out will decrease with increasing sampling height of the precipitation. As an example, the age determined in March 1959 from the strontium-89/strontium-90 ratio was 170 days at 0 m. and 148 days at 123 m. above ground-level.

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[1] Herbst, W., Langenderff, H., Philipp, K., and Sommermeyer, K., "Untersuchungen über Radioaktivität der Vegetation, Atomkernenergie", Heft 10, 369 (Oct. 1957).
[2] Small, S. H., "Some Aspects of the Use of Naturally Radioactive Isotopes as Tracers in Meteorology", *IR-F* 307.
[3] Aarkrog, A., and Lippert, J., Risö Report No. 9 (June 1960).
[4] Möller, H. Bjerrum, and Jensen, K., Risö Report No. 9 (February 1960).
[5] Osmond, R. G., Proishaft, A. G., and Warricher, J. B., *A.E.R.E., C/R* 2166 (August 1957).
[6] Aarkrog, A., and Lippert, J., Risö Report No. 14 (June 1960).
[7] Chamberlain, A. C., *A.E.R.E., HP/R* 1261 (1955).

Printed in Great Britain by Fisher, Knight & Co., Ltd., St. Albans.

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Variation in the Ratio of Strontium-89 to Strontium-90 in Precipitation Samples with Sampling Height

It was shown that during March–December 1959 (ref. 1) the specific activities of fission products from fall-out in precipitation increased with sampling height. Some evidence was also found for an increase in the ratio $^{89}Sr/^{90}Sr$ with sampling height, but this increase was not significant ($P \sim 90$ per cent).

After the resumption of large-scale nuclear testing in September 1961, it became possible to repeat the experiment over a longer period. The ^{89}Sr ^{90}Sr ratios were determined in precipitation collected at eight different heights in the meteorological mast at Risö after the principles used in 1959 (ref. 1).

Table 1 shows the results for the 24-month period: October 1961–September 1963.

The $^{89}Sr/^{90}Sr$ ratios in Table 1 were treated as follows. The ratios were converted to the logarithms, because the figures were logarithmically distributed. The converted ratios for each month were divided by the corresponding (converted) mean ratio for the month. The purpose of this operation was to eliminate the effect of the time variation of the $^{89}Sr/^{90}Sr$ ratios. The months were separated into two groups, one with high variance (I) and one with low (II). This was necessary for the performance of a regression analysis, where the differences in the variances have to be insignificant. The two groups of months were each sub-divided into two sub-groups, one representing 'dry' month (d), that is, months with less than 25 mm precipitation and another representing 'wet' month (w), that is, months with 25 mm and more precipitation (Table 1).

The following regression lines were calculated:

$$y = 1 + 0{\cdot}1040 \times 10^{-2}\,(x - 46{\cdot}55) \qquad \text{(Id)}$$

$$y = 1 - 0{\cdot}4452 \times 10^{-2}\,(x - 52{\cdot}13) \qquad \text{(Iw)}$$

$$y = 1 + 0{\cdot}4050 \times 10^{-2}\,(x - 52{\cdot}13) \qquad \text{(IId)}$$

$$y = 1 + 0{\cdot}6857 \times 10^{-2}\,(x - 52{\cdot}39) \qquad \text{(IIw)}$$

(where y is the transformed $^{89}Sr/^{90}Sr$ ratio, and x is the sampling height).

Table 1. Ratio of strontium-90 to strontium-89 in precipitation collected at eight different heights in the Finish Defence, 1961-September, 1962

Height (m) Month	1	7	25	30	54	72	96	152	Mean	Min precipitation band group
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

Figures in brackets are missing values calculated by the method of least squares. These figures are omitted in the regression analysis.

Table 2. COMPARISON OF THE LINES IN GROUP I ANALYSIS OF VARIANCE

Variation	Sum of squares of deviations	Degrees of freedom	Variance	Test
about lines	0·4147	57	$0·7275 \times 10^{-2}$	
between slopes	$0·5045 \times 10^{-1}$	1	$0·5045 \times 10^{-1}$	6·04

Table 3. COMPARISON OF THE LINES IN GROUP II. ANALYSIS OF VARIANCE

Variation	Sum of squares of deviations	Degrees of freedom	Variance	Test
about lines	$0·7378 \times 10^{-1}$	114	$0·6472 \times 10^{-3}$	
between slopes	$0·4952 \times 10^{-2}$	1	$0·4952 \times 10^{-2}$	7·65

An analysis of variance showed significant differences between the slopes for dry and wet months for both group I ($P \sim 99$ per cent) and group II ($P > 99$ per cent) (Tables 2 and 3).

Hence, it is evident that in 'dry' months the slope was significantly steeper than in the 'wet' months, that is, the $^{89}Sr/^{90}Sr$ ratios in 'dry' months were significantly higher in the upper precipitation collectors than in the lower. An explanation for this phenomenon has been proposed earlier[1]: it is assumed that the dry deposit, which yields the higher concentrations in the upper precipitation collectors, is of greater particle-size than the deposit from rain. Larger and heavier particles will be deposited more readily than fine particles, and consequently the coarse particles will consist of younger fission products (greater $^{89}Sr/^{90}Sr$ ratio) than the fine ones.

I thank Mr. Heinz Hansen for his advice concerning the statistics.

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[1] Aarkrog, A., *Nature*, **188**, 482 (1960).

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Europium-155 in Debris from Nuclear Weapons

Abstract. *The lithium-drifted germanium detector enables determination of europium-155 on a routine basis in environmental samples contaminated with debris from nuclear weapons. From measurements of europium-155, cesium-144, and strontium-90 in air filters collected between 1961 and 1966, the yield of europium-155 from weapons was estimated at 1400 atoms per 10^6 fissions, which is close to the yield of europium-155 from fast fission of uranium-238.*

In studies of circulation processes in nature, such as in meteorology, hydrography, and ecology, debris from the testing of nuclear weapons has been widely used. The long-lived nuclides Sr^{90} and Cs^{137} are most commonly employed in these investigations, but radionuclides of medium-long life, such as Ce^{144}, Ru^{106}, Sb^{125}, and Pm^{147}, also have found some applications. The great spectral-resolving power of the lithium-drifted germanium detector has made it possible to include 1.7-year Eu^{155} in this family of useful environmental tracers (*1*).

Europium-155 is a fission product having a thermal neutron fission yield, in U^{235}, of $326:10^6$ (atoms:fission) (*2*). In debris from nuclear weapons, Eu^{155} was detected for the first time in 1957 in soil on Rongelap Atoll (*3*); later it was measured in global fallout in samples of rain water (*4, 5*), dust (*5*), lichens (*1*), plankton and sea water (*6*), and marine sediments (*7*). Our aim has been to follow the concentration of Eu^{155} in ground-level air for a longer period, and, from these measurements and simultaneous determinations of Ce^{144} and Sr^{90}, to estimate the weapon yield of Eu^{155}.

Since 1961, air samples have been collected at ground level at Risö, Denmark, by means of a 7.5-hp centrifugal pump handling air at about 10^6 m^3/-month. The debris was collected on two Whatman-GF/A glass-fiber filters (each 56 × 48 cm) having a filter efficiency of about 100 percent (*8*); normally they were changed twice weekly. A monthly portion of 10 g of pressed filter, representing about 30,000 m^3 of air, was measured on a 2.3-cm^3 lithium-drifted germanium detector (*1*) in concert with a TMC-256 multichannel pulse-height analyzer; the counter was operated as a Compton-rejection spectrometer (*9*). Most of the samples were counted for 1000 minutes. The 105-kev photo peak was used for the calculations of Eu^{155}; the 134-kev peak, for Ce^{144}. All samples were corrected for decay back to the middle of the month of sampling.

The maximum of the Eu^{155} determinations (Table 1) (*10*) occurred, as for other long-lived, bomb-produced nuclides (*11*), in June 1963 (Eu^{155}, 0.025 pc/m^3). The Chinese nuclear tests, made since 1964, have resulted in transient increases in the atmospheric concentrations of Eu^{155} during the first months after the explosions.

Along with the Eu^{155} measurements, Ce^{144} was determined; Fig. 1 shows the Ce^{144}:Eu^{155} ratios and the decay curves for the mean ratios on 15 October 1961 and 15 November 1962. Strontium-90 was determined by radiochemistry from paper-filter samples from another air sampler at Risö (*11*); the Eu^{155}:Sr^{90} ratios calculated from these measurements appear in Fig. 1 (bottom).

For estimation of the mean ratios at formation, two periods (indicated by the solid decay curves in Fig. 1) were selected: (i) from November 1961 to August 1962—from the end of the 1961 series to the month when the fresh fallout from the 1962 series became important; and (ii) from after the 1962 series (from January 1963) until September 1964—the month before the resumption of atmospheric tests by China. The ratios from these two periods were referred to 15 October 1961 and 15 November 1962, respectively, the reference dates being the estimated mean dates for the 1961 and 1962 series (*12*).

Table 2 shows the mean ratios, and the standard errors of the means, calculated for the two periods. It is not surprising that the relative errors of the Eu^{155}:Sr^{90} mean ratios are greater than the errors of the Ce^{144}:Eu^{155} ratios when one considers that Sr^{90} and Eu^{155} were determined from different samples taken by different samplers, whereas Ce^{144} was determined simultaneously with Eu^{155} from the same samples.

Pre-1961 debris was disregarded in the following calculations because the contribution in 1962 from this old debris was less than 10 percent; in 1963–64, less than 5 percent (*13*). It is estimated that about 80 percent of the debris in 1963–64 came from the 1962 series, the remainder coming from the 1961 tests (*13, 14*). Thus the ratios on 15 November 1962 were corrected (Table 2) by omitting the contributions of debris from the 1961 series (*15*).

While the Ce^{144}:Eu^{155} ratios on the two dates of formation are in good agreement, the difference between the Eu^{155}:Sr^{90} ratios is obvious; it may merely result from the greater error of the Eu^{155}:Sr^{90} ratios, but it may reflect a real difference between the ratios from the 1961 and 1962 test series—perhaps due to fractionation. This phenomenon is less likely to be important for the Ce^{144}:Eu^{155} ratio, as the precursors of these nuclides may be less volatile than the precursors of Sr^{90} (*16*).

Table 1. Europium-155 in air samples (pc/10^3 m^3). The relative S.D. of a single determination was 23 percent (*10*).

Month	1961	1962	1963	1964	1965	1966
January		3.6	10.0	4.9	1.1	0.27
February		4.6		5.6	1.0	.43
March		5.1	19.7	8.5	1.9	.36
April		4.2	17.2	12.1	1.8	.44
May		6.4	18.2	9.5	5.2	.59
June		11.8	25.2	10.5	1.3	.60
July	0.11	9.4	15.5	3.7	1.3	.36
August	.55	4.9	7.6	4.0	0.66	.28
September	1.1	3.7	7.1	2.8	1.3	.21
October	1.9	3.5	8.1	0.83	1.0	.22
November	3.1	4.7	4.1	1.6	0.41	.16
December	5.8	5.7	3.6	1.2	.24	.06

Table 2. Mean ratios of formation of Ce^{144} and Eu^{155} and of Eu^{155} and Sr^{90} in debris from nuclear weapons.

Date	Ce^{144} : Eu^{155}	Eu^{155} : Sr^{90}
15 Oct. 1961	68 (S.E., 3)	0.84 (S.E., 0.13)
15 Nov. 1962	71 (S.E., 2)	.48 (S.E., 0.02)
Corrected for 1961 debris		
15 Nov. 1962	74	0.47
Mean ratios for 1961 and 1962 debris	71 ± 3	0.66 ± 0.19

The mean ratios in Table 2 are rather close to the ratios found for fast fission in U^{238} [Ce^{144}:Eu^{155}, 64.4; Eu^{155}:Sr^{90}, 0.78 (*2*)]. Kuroda *et al.* (*4*) found a Eu^{155}:Sr^{90} ratio of 0.24 in precipitation on 16 October 1964; this ratio fits the decay curve in Fig. 1 excellently.

Harley *et al.* (*17*) have measured the weapon yields for Ce^{144} and Sr^{90} at 46,900 and 35,000 atoms per 10^6 fissions, respectively. From these determinations and the mean ratios in Table 2, the weapon yield of Eu^{155} is calculated at $1400/10^6$ (atoms/fissions). As 1 kton of fission corresponds to 1.45 $\times$ 10^{23} fissions (*17*), production of Eu^{155} by nuclear weapons is estimated at 72.6 kc per megaton of fission. The total fission yield of the 1961–62 test series was 101 Mtons (*18*), so that the total production of Eu^{155} in these tests was 7.3 Mc.

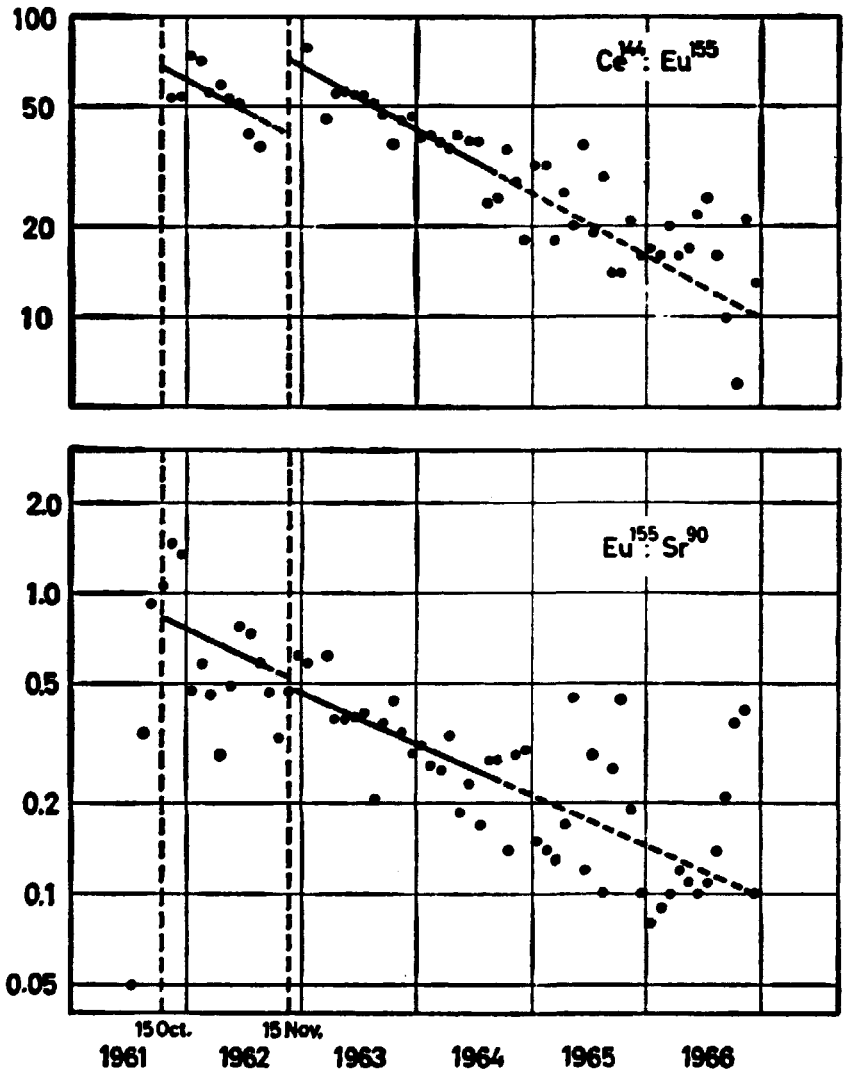

Fig. 1. Ratios Ce^{144}:Eu^{155} and Eu^{155}:Sr^{90}, with decay curves, in air-filter samples.

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References and Notes

1. J. Lippert, "Some applications for semiconductor detectors in health physics," before meeting Nordic Soc. Radiation Protection, Stockholm, Sweden, Feb. 1966.
2. L. E. Weaver, P. O. Strom, P. A. Killeen, *USNRDL-TR-633* (March 1963).
3. R. F. Palumbo and F. G. Lowman, *UWFL-56* (1958).
4. P. K. Kuroda *et al.*, *Science* **147**, 1284 (1965).
5. H. Klös, R. May, H. Schneider, *Z. Naturforsch.* **21a**, 363 (1966).
6. B. Schreiber, E. Cerrai, C. Triulzi, L. Tassi-Pelati, *Rend. Ist. Lombardo Sci. Letter* **B98**, 143 (1964).
7. V. E. Noshkin and V. T. Bowen, "Vertical distribution of individual radioisotopes from fallout in marine sediment cores," before Amer. Chem. Soc. Symp. on Forefronts in Terrestrial Geochemistry, Atlantic City, N.J., Sept. 1965.
8. L. B. Lockhart, Jr., and R. L. Patterson, Jr., *NRL Rept. 6054* (1964).
9. Y. Sever and J. Lippert, *Nucl. Instr. Methods* **33**, 347 (1965).
10. The amount of air in the Feb. 1963 sample is unknown, so that exact values of the Eu^{155} concentration and the Eu^{155}:Sr^{90} ratio for this month are not available. About one-third of the figures in Table 1 rely on double determination. Europium-155 was identified in the June 1963 sample by radiochemistry. The rare earths were isolated by ion exchange and europium was reduced by zinc-amalgam to Eu^{++}, thus preventing it from precipitating as the hydroxide with gaseous NH_3 along with the other rare earths.
11. A. Aarkrog and J. Lippert, *Risö Rept. 41, 63, 85, 107, 131* (1962–66).
12. L. P. Salter, in *Radioactive Fallout from Nuclear Weapons Tests*, A. W. Klement, Ed. (AEC, Washington, D.C., 1965), p. 411.
13. P. F. Gustafsson, S. S. Brar, S. E. Muniak, *ibid.*, p. 504.
14. D. H. Peirson and R. S. Cambray, *Nature* **205**, 433 (1965).
15. The contributions of Eu^{155} and Ce^{144} in 1963–64 from the 1961 series were corrected for decay and were thus less than the 20 percent taken as the Sr^{90} contribution from the 1961 tests.
16. E. C. Freiling, *Science* **133**, 1991 (1961); ——— and M. A. Kay, *Nature* **209**, 236 (1966).
17. N. Harley, I. Fisenne, L. D. Y. Ong, J. Harley, in *HASL-164* (1 Oct. 1965), p. 251.
18. U.S. Federal Radiation Council, *Rept. 4* (May 1963).
19. We thank Miss Bodil Lassen for the Eu^{155} determinations.

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RADIOECOLOGICAL INVESTIGATIONS OF PLUTONIUM IN AN ARCTIC MARINE ENVIRONMENT

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Abstract—Samples of the human food chain at Thule, Greenland, were collected during the summer of 1968, after the nuclear weapon incident in January. As was to be expected from the increased plutonium levels in bottom sediments, the highest animal levels were found in bivalves, crustacea, polychaeta, and echinodermata. The levels in these bottom animals were on the average two orders of magnitude as high as the fall-out background, in a few cases four orders of magnitude. Fish from the bottom water also showed an increased plutonium content whereas sea weed, plankton, sea birds, seals, and walruses did not differ significantly from the fall-out background. The plutonium concentration in sea water was twice the fall-out background. No samples displayed plutonium levels that were considered hazardous to man or higher animals in the Thule district or in any other part of Greenland.

INTRODUCTION

ON 21 JANUARY 1968, a B-52 aeroplane from the U.S. Strategic Air Command crashed on the ice in Bylot Sound 11 km west of Thule Air Base, Greenland (Fig. 1). By the impact with the ice the chemical explosive in the four unarmed nuclear weapons carried by the aircraft detonated, and some kilogrammes of plutonium were released to the environment.

Most of the contamination was confined within a dropshaped area of an approximate size of 700 × 150 m stretching southward from the point of impact. Bomb debris, wreckage from the plane and the surface layer of the contaminated snow in the drop-shaped area were removed by the U.S. Air force during the first few months after the accident and shipped in the autumn to the Savannah River plant in the United States.

Preliminary surveys

Around the point of impact the ice was broken, and it was just possible that a few pieces of debris had gone through the ice and thus caused a contamination of the marine environment. From holes cut through the approx. 1 m thick ice, bottom sediments and plankton were collected in the first weeks after the accident. In a few cases these samples showed measurable plutonium levels. It could not, however, be ruled out that the samples had been contaminated by the surface snow during the sampling. Besides the marine samples a number of arctic foxes were analysed for plutonium; none of them showed significant levels.

By the middle of February more than one hundred samples of surface snow had been collected outside the drop-shaped area. The extra-sital contamination was found to be concentrated in two sectors, one extending southward to Narssarssuk and one westward to the south-west coast of Saunders Island. The maximum plutonium deposition measured was 40 pCi/cm² (~0.4 Ci/km²), and the geometric mean of all samples was 0.4 pCi/cm² (~4 mCi/km²). From this snow survey it was estimated that the contamination of the Thule environment outside the impact area was few curies of ^{239}Pu.

Whether plutonium had been spread to more distant locations and especially to the neighbouring Dundas village was checked by the sampling of surface snow from 25 locations around the coasts of Greenland, from station Nord in the north-eastern corner of Greenland to Dundas in the north-western corner. None of these samples showed any indication of surplus ^{239}Pu activity.

HASL[1] carries on a routine collection of air samples at the Thule Geopol Station, which

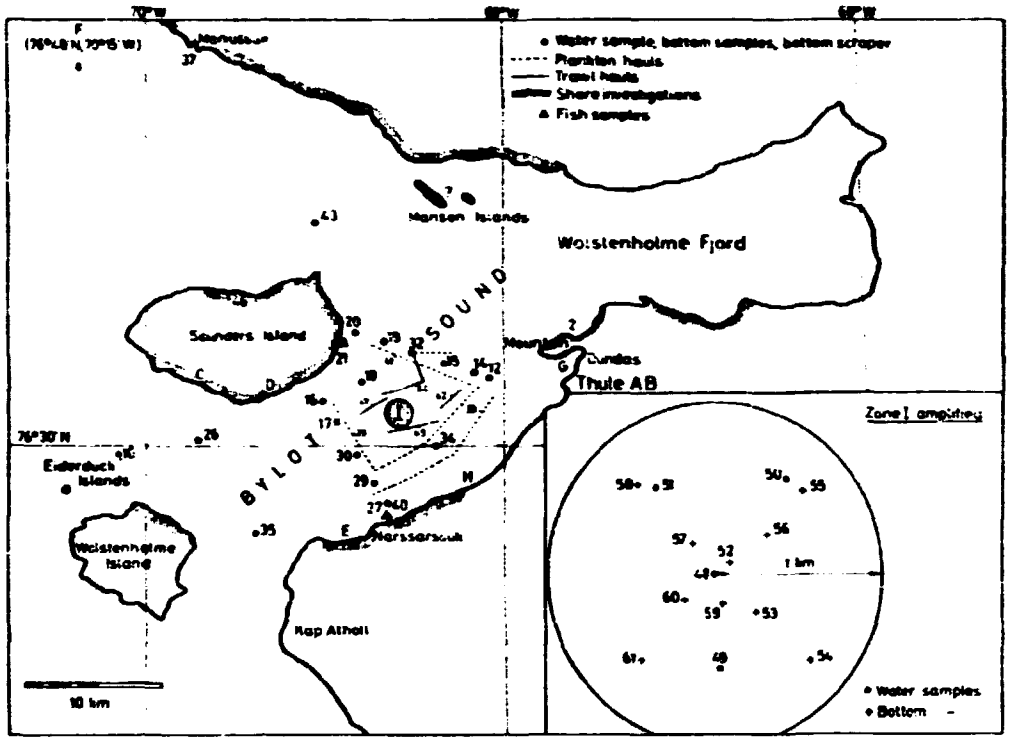

FIG. 1. The sampling area at Thule, Greenland. Zone I has its centre at the point of impact.

is located a few miles east of the air base at a 259 m mountain hill. In January 1968 the ^{239}Pu level was 0.067 pCi ^{239}Pu/10^3 m^3 at Thule, which was somewhat higher than at the 12 other northern stations (0.027 (1 S.D. = 0.013)) in the HASL network. In December 1967 and February 1968 the Thule levels were 0.019 and 0.045 pCi ^{239}Pu/10^3 m^3 respectively. The ^{239}Pu/^{90}Sr ratio in January was 0.056 at Thule, while at the other stations in the northern hemisphere it was 0.021 (1 S.D.:0.002), and the ^{238}Pu/^{239}Pu ratios were respectively 0.22 and 0.54 (1 S.D.: 0.23). Thus there is an indication of a surplus ^{239}Pu activity in January 1968 at Thule from the accident, although the increase is modest.

The cleaning up of the crash site was very efficient, and it was estimated that only approx. 25 Ci (±50%) remained on the ice. When the ice in June–July broke up in Bylot Sound, it was observed that the crash area drifted northward. However, no observations revealed where and when the contaminated ice was transferred into the sea. A radioecological study when Bylot Sound was navigable was thus indicated, to obtain information on the fate of the released plutonium in the biosphere.

Preaccidental plutonium levels

Since 1959 HASL[1] has measured the ^{90}Sr fall-out at Thule. The accumulated level for the period 1959–68 was 9.2 mCi/km^2; the deposit prior to 1959 was estimated at 2.6 mCi ^{90}Sr/km^2 (from the ^{90}Sr inventory in 1958 in the northern hemisphere[5]). Hence the total ^{90}Sr deposit at Thule until 1968 was approx. 12 mCi ^{90}Sr/km^2 (not corrected for decay).

Several authors[1–4] have estimated the ^{239}Pu/^{90}Sr ratio in fall-out (cf. Table 1). In the following a ratio of 0.023 is applied. The accumulated ^{239}Pu fall-out at Thule was thus estimated at 12 × 0.023 mCi/km^2 = 0.28 ~ 0.3 mCi/km^2. The area of Bylot Sound is about 300 km^2. Hence the total amount of plutonium in the sound prior to the accident was 0.1 Ci ~ 1–2 g.

Table 1. $^{239}Pu/^{90}Sr$ *ratios in fall-out*

Sample type	Sample period	Sample location	Ratio	Author and reference
Precipitation	1958–1966	Tokyo, Japan	0.016	MIYAKE *et al.*[2]
Ground-level air	1961–1965	Italy	0.022	BORTOLI *et al.*[3]
Ground-level air	1963–1968	U.K.	0.023	CAMBRAY *et al.*[4]
Ground-level air	1965–1968	New York	0.024	VOLCHOCK[1]

The accident was thus expected to have produced an average increase of the plutonium burden at Thule by approx. two orders of magnitude, provided the main part of the contamination was confined to Bylot Sound.

MATERIAL AND METHODS

The Thule environment

Thule is located in the north-western corner of Greenland at about 76°30'N and 69°W. The climate is arctic: during the winter (December–March) the mean temperature is −22°C, and in the summer months July–August it is +4°C. For three months, beginning in November, the sun remains below the horizon, while for three and a half months during the summer the "midnight sun" shines. The climate is dry with an annual mean precipitation of less than 100 mm.[1]

Bylot Sound is open from July to September. Between Saunders Island and Wolstenholme Island the ice may, however, disappear already in April. The sea water has a mean temperature of 0°C or a little less. The mean depth of Bylot Sound is 100 m: in the impact area the depth was nearly 200 m.

The Eskimos

The local population have two settlements along the coasts of Bylot Sound and Wolstenholme Fjord: Maniussak with 6–8 families and Narssarssuk with two families. Within a distance of 25 km from the crash site about 60 Eskimos are living. The total population of Greenlanders in North Greenland is approx. 600.

The food chain at Thule[6] (*Fig.* 2)

Phytoplankton is produced in appreciable quantities during the light time of the year. It can be extremely abundant, especially near the bird cliffs of Saunders Island, where the sea is fertilized by the guano from the birds. The phytoplankton is eaten by zooplankton animals as well as many of the bottom animals.

Zooplankton is found the year round from the bottom to the surface. In winter it is most abundant near the bottom and consists mainly of *sagitta*, *aglanta* and *calanus*. In summer, species of *medusas*, *stenophores*, *pteropods*, *euphasians*, *copepods*, *amphipods*, *mysis* and fish larvae are found. The zooplankton is eaten by many lower sea animals, but also by fish and birds such as the polar cod (*Boreogadus saida*) and the little auk (*Plotus alle*).

Bivalves, Crustacea and other bottom animals. The sea bottom in Bylot Sound consists of fine clay, in some places near the coast mixed with sand and pebbles. In the western part of the sound the bottom is rocky. Bivalves (*cardium*, *macoma*,

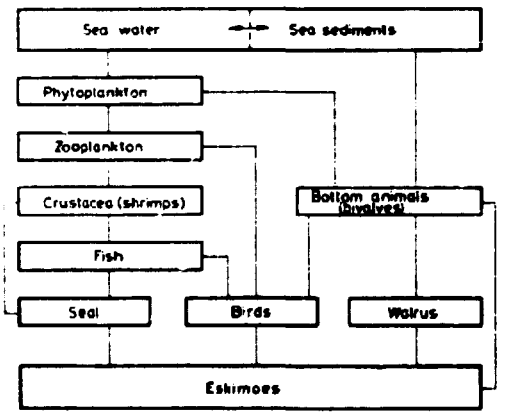

FIG. 2. Food chains in an arctic, marine environment.

chlamys, *hiatella* and *astarte*), snails (*buccinum*), worms (*polychaeta*), crustacea (shrimps), *echiodermata* (starfish, sea urchins, etc.) and corals are abundant, their total density amounting at certain locations to up to 450 g/m^2.

The sea bottom bivalves are eaten by the walruses and the eiders. The Eskimos occasionally collect mussels (*Mytilus edulis*) along the beach during ebb-tide; they also eat bivalves (*cardium*, *mya* and *hiatella*) from the stomach of the walrus.

Crustaceans are eaten by ringed seal, eider and the Greenland halibut.

Fish. The polar cod is the most abundant fish at Thule. It is eaten by the seals and whales and by Brünnick's guillemot. At the bottom, the families *liparidae*, *lycodidae* and the Greenland halibut (*Reinhardtius hippoglossoides*) are found. A few halibuts are caught from the ice during winter and north of the Thule area. Shark (*Somnius microcephalus*) and sea scorpion (*Cottus scorpius*) are found along the coasts and caught for dog food. Commercial fishing does not take place in the Thule district.

Walrus. A few hundred walruses are caught every year in the Thule district. The meat, liver, kidneys, and heart are occasionally consumed by humans, but are primarily used for dog food. The walrus can take its food from depths of as much as 80 m.

Other Seals. The bearded seal (*Erignathus barbatus*) and the ringed seal (*Phoca hispida*) are common in both summer and winter, while the harp seal (*Phoca groenlandica*) is found mainly during early summer. The seal is the most important hunting animal for the Eskimos, who eat the meat, liver, heart and kidneys.

Birds. The little auk (*Plotus alle*) breeds in millions in the mountains from Cape Atholl and southward to Melville Bay. It serves as the chief food of the arctic fox during summer. In May and June the Eskimos catch the little auk in nets and collect its eggs, in August they catch the young ones before they can fly.

Brünnich's guillemot (*Uria lomvia*) breeds on the western side of Saunders Island. The eggs are collected in the bird cliffs, and the birds are hunted in June and July.

The black guillemot (*Cephus grylle*) breeds in the scree along all the coasts. It is shot especially in spring along the crevices in the ice. The glaucous gull (*Larus hyperboreus*) and the kittiwake (*Rissa tridactyla*) breed in many places, e.g. on the cliffs of Saunders Island. They are shot in a modest number during summer.

The eider (*Somateria mollisima*) breeds on the Eiderduck Islands and the Manson Islands and in other places. Some eider hunts are carried out during summer, but collecting of eggs and eiderdown is of greater importance.

The raven (*Corvus corax*) is common everywhere in both, summer and winter. It eats nearly anything that is edible. Many young birds are shot at the end of the summer and caught in fox-traps.

Other animals. The white whale (*Delphinapterus leucas*) and the narwhale (*Monodon monoceros*) are found in the Thule district, but the most important hunting ground for the whales is Inglefield Fjord 60 km north of Bylot Sound.

The arctic fox (*Alopex lagopus*) is common throughout the district, 1000–1500 being shot or caught every year. The meat is sometimes eaten, but the animals are primarily hunted for their skins.

The polar bear (*Ursus maritimus*) is an occasional guest in the area. The bear feeds on the ringed seal. Between 20 and 25 bears are shot annually in North Greenland. The meat is eaten.

The hare (*Lepus variabilis*) eats grass and other vegetation. It is shot in small numbers.

The sampling programme

The samples were to be collected in three zones. Zone I was a circular area with its centre at the point of impact and a radius of 1 km. Zone II consisted of the remaining part of Bylot Sound and Wolstenholme Fjord with the islands and coasts shown in Fig. 1. Zone III was a control zone, situated outside the area shown in Fig. 1, and included Qanaq about 100 km north of Thule AB. Samples were first collected in zone II, then in III and finally in I; by this order it was ensured that possible hot samples from zone I did not contaminate the samples from zones II and III.

Sea water, bottom sediments, zooplankton (shrimps) and bivalves were considered the most important samples because the levels in them would be decisive for the levels in the other links of the food chain (Fig. 2). The

secondary samples were excretions or intestinal contents from seals and birds, besides seaweed and fish. Finally human urine samples were collected from Narssarssuk.

The scientific expedition consisted of one zoologist, one marine biologist, one hydrographer, one lichenologist, two health physicists, and two assistants for the sampling. The investigations began in the last week of July and were in all essentials finished by the end of August. By then more than 150 samples had been collected for plutonium analysis.

The marine sampling was carried out from the good ship "Aglanta," the former Greenland fisheries and marine research vessel. The sampling from the shores was carried out partly by motor boat, partly by USAF helicopters.

The samples were in many cases collected with specially constructed equipment. Bottom sediments were thus collected by means of a 0.1 m^2 bottom sampler, which collected only the uppermost centimetre of the sea floor. Shrimps and polar cods were caught in a shrimp trawl, and sea water was collected from different depths with a 100 l. water sampler. Bivalves were caught with an ordinary triangle dredge.

Sample treatment

The biological samples were kept at −10°C until the processing in the laboratory could take place. The solid samples were ashed at 600°C, and ^{236}Pu spike and Fe-scavenger were added to 2–10 g ash for one analysis. Potassium pyrosulphate was mixed with the ash in the ratio 3:1, and the mixture was heated to melting. By this treatment the oxidation step of plutonium was adjusted to +4 and all plutonium brought into a soluble form. The plutonium was precipitated as hydroxide along with the iron scavenger. Then the mixture of iron and plutonium was ion exchanged on an anion resin, which separated the iron from the plutonium. Finally the plutonium solution was subjected to extraction with 1-phenyl-3-methyl-4-benzoyl-pyrazolone-5 in xylene. The organic phase, which contained the plutonium, was evaporated on a stainless-steel planchette, ignited and counted on a silicon surface barrier counter connected with a 256-channel analyser. The detailed chemical procedure has been described elsewhere.[16]

The sensitivity of the procedure, defined as three times the activity found in a blank, was better than 1 fCi/l. (10^{-15} Ci/l.) sea water for a 50 l. sample (the normal size of sea water samples for analysis) and approx. 10 fCi/g ash for a 4 g ashed sample. Most samples were counted for 4 × 1000 min and from the four countings the S.E. of the results was estimated. If the error was greater than 33%, the sample was not considered significantly different from zero background. Such samples are denoted by a B in the tables. Samples with errors between 20 and 33% are denoted by an A. Some samples were analysed more than once and in those cases ±1 S.E. was indicated.

RESULTS AND DISCUSSION

Sea water

It is evident (cf. Tables 2–5) that the plutonium levels in sea water were so low that in several cases it was impossible to demonstrate whether the levels were at all above the zero background. However, it seems probable that water from Bylot Sound contained significantly more plutonium than sea water collected from localities outside Thule (zone III). The median level of ^{239}Pu in sea water from zones I and II was approx. twice the fall-out background in sea water, which from Tables 4 and 5 was estimated at 2 fCi/l.

Pillai *et al.*[17] have earlier measured the ^{239}Pu levels in Pacific sea water, and in surface sea water collected near the Californian coast in January–April 1964 they found 0.35–0.39 fCi/l. On the high seas, respectively 1500 and 2500 km from the coast, they found 1.9 and 3.0 fCi/l.; the $^{239}Pu/^{90}Sr$ ratio of the sample taken 2500 km from the coast was 0.008, i.e. somewhat lower than in fall-out (cf. Table 1).

The present estimate of the fall-out background in ocean water is in reasonable agreement with the observations of Pillai *et al.* as far as their results from the open Pacific are concerned. Their levels in coastal waters are, however, definitely lower than the present results, although there are some indications of lower plutonium levels also in the Danish inner waters, whereas the ^{90}Sr levels are higher in the inner waters than in the open sea. Thus the median level of the $^{239}Pu/^{90}Sr$ ratio for Greenland sea water outside Thule (cf. Table 4) was 0.018 (i.e.

Table 2. ^{239}Pu levels in sea water collected in August 1968 in zone I, at Thule, Greenland

Location (cf. Fig. 1)	Sample depth in metres	Sampling date	Salinity in ‰	(fCi/L)	$^{239}Pu/^{90}Sr$
48	2	Aug. 21		4.8 ± 0.5	
48	180	Aug. 21		9.1 ± 1.3	
49	2	Aug. 21		4.3	
49	195	Aug. 21		2.1	
49	195	Aug. 21		4.9*	
50	2	Aug. 21		4.8	
50	175	Aug. 21		1.7A	
50	175	Aug. 21		3.4*	
51	2	Aug. 21	31.5	11.7*	0.072
51	178	Aug. 21	33.4	4.4*	0.034
Median				4.5	

* The samples were filtered through a 1 μ millipore filter before the analysis. The filters were measured, but showed levels, which were not significantly different from filters used for the filtration of similar volumes of distilled water.

Table 3. ^{239}Pu levels in sea water collected in August 1968 in zone II at Thule, Greenland

Location (cf. Fig. 1)	Sample depth (m)	Sampling date	Salinity in ‰	(fCi/l.)	$^{239}Pu/^{90}Sr$
12	85	Aug. 6		5.7	
11	225	Aug. 6		76	
15	99	Aug. 6		1.5	
16	88	Aug. 6		8.7	
17	166	Aug. 6		7.0	
18	90	Aug. 6		3.2	
30	254	Aug. 11		46	
29	95	Aug. 11		2.7A	
27	30	Aug. 11		67	
34	130	Aug. 15		2.9	
35	260	Aug. 15	35.0	6.5	0.050
26	34	Aug. 9	33.6	3.0A	0.025
43	100	Aug. 20	33.5	6.6	0.049
Median				6.5	

Samples 32, 19 and 20 were lost during the analysis

Table 4. ^{239}Pu in Greenland sea water outside Thule (zone III) collected in August 1968

Location	Position N	Position W	Sample depth (m)	Salinity in ‰	(fCi/l.)	$^{239}Pu/^{90}Sr$
Qanaq	77°25′	69°29′	2	28.3	3.2	0.018
Qanaq	77°25′	69°29′	200	37.0	2.9 ± 0.6	0.039
Godhavn	~69°	~54°	surface	33.2	3.8	0.025
Godthåb	~64°	~52°	surface	32.4	2.5	0.014
Prins Chr. Sund	~60°	~45°	surface	27.5	26	0.115
Angmagssalik	~65°	~38°	surface	23.9	~1B	0.004
Danmarkshavn	~77°	~19°	surface	29.8	4.1	0.014
Median					3.2	0.018

Table 5. ^{239}Pu in sea water from the inner Danish waters collected in October 1968 by the "DANA"

Location	Position N	Position E	Sample depth (m)	Sampling date	Salinity in ‰	(fCi/l.)	$^{239}Pu/^{90}Sr$
The Sound	55°56′	12°39′	0	Oct. 10	19.2	2.1A	0.003
The Sound	55°56′	12°39′	32	Oct. 10	35.7	~0.2B	0.001
The Categat	56°07′	11°07′	0	Oct. 11	21.3	0.7B	0.002
The Categat	56°07′	11°07′	32	Oct. 11	33.6	1.6	0.004
The Great Belt	55°21′	11°02′	0	Oct. 11	20.1	1.1	0.003
The Great Belt	55°21′	11°02′	31	Oct. 11	33.3	2.9A	0.012
The Baltic	54°28′	12°15′	0	Oct. 11	12.6	3.6	0.005
The Baltic	54°28′	12°15′	23	Oct. 11	20.7	1.7	0.003
The Baltic	55°07′	12°55′	0	Oct. 12	7.6	1.3A	0.002
The Categat	57°48′	10°42′	0	Oct. 26	34.6	~0.1B	0.001
The Categat	57°48′	10°42′	70	Oct. 26	35.6	~1.2B	0.005
Median						1.6	0.003

close to the ratio in fall-out), while for Danish inner waters (cf. Table 5) it was only 0.003.

It was not possible to show any significant difference between the plutonium levels in surface- and bottom-water, although in certain cases the ^{90}Sr levels differed markedly (cf. e.g. the samples from Qanaq in Table 4). This is in agreement with observations made by Bowen and Sugihara on lanthanides[8] (^{144}Ce and ^{147}Pm). They supposed that these lanthanides were removed on particles sinking at a rate of about 100 m or more per month.

Let us suppose the mean concentration to be 2 fCi ^{239}Pu per litre sea water in Bylot Sound owing to the accident. The total volume of the sound is about $300 \cdot 10^6 \cdot 100 \cdot 10 = 3 \cdot 10^{13}$ l.; hence the total ^{239}Pu content from the accident dissolved in the sea should be ~0.06 Ci. A number of samples (cf. Table 2) were filtered through a 1 μ millipore filter before the analysis, and filtrate and filters were analysed separately. These analyses gave no unambiguous indications of significant amounts of particulate activity in the water samples. However, it is believed that the few samples that showed relatively high levels (>10 fCi ^{239}Pu/l.) contained particulate activity, probably particles stirred up from the bottom during the sampling. Most samples were collected near the bottom (cf. the tables) as it was believed that the highest concentrations would be found there.

Sea bottom sediments

The plutonium from the accident was present as PuO_2 with a median particle size of 2 μ (S.D. 1.7).[9] Many of the plutonium particles were attached to inactive material such as fuel, burned plastic, etc., with a median diameter of the inert particles 4–5 times that of the plutonium particles. Sedimentation experiments[9] on melted ice cores showed that 85–95% of the debris and associated plutonium oxide sank immediately in water, and that only 1% was suspended as fine particles in the water phase. The remaining part was retained on the surface together with a thin film of fine, carbonized material. It was thus to be expected that most of the plutonium that had not been removed by the decontamination of the accident area was to be found on the sea bottom. It was, however, not certain that the main part of the activity was concentrated below the point of impact in zone I as appreciable amounts might have drifted away on the ice before sinking to the bottom.

Table 6 and Fig. 3 demonstrates that zone I showed higher levels than zone II, but it is evident that high levels could also be encountered outside zone I (cf. locations 30 and 17). Let us suppose that most of the activity had been concentrated within an elliptic area stretching from locations ?? and 14 through zone I to locations 17 and 30. The median deposition in

Table 6. ^{239}Pu levels in bottom sediments collected in August 1968 at Thule, Greenland

	Location (cf. Fig. 1)	Sampling date	pCi/g ash	mCi/m² or mCi/km²	$^{238}Pu/^{239}Pu$
Zone II	12	Aug. 12	0.10	0.19	
	14	Aug. 12	1.3	3.9	
	17	Aug. 10	2.0 ± 0.3	9.7 ± 1.4	
	19	Aug. 13	1.1	0.84	
	30	Aug. 11	16 ± 5	71 ± 20	0.020
	32	Aug. 13	1.3 ± 0.2	5.5 ± 0.9	0.014
	45	Aug. 20	0.13 ± 0.01	0.92 ± 0.04	
	Median		1.3	3.9	
Zone I	55	Aug. 22	7.0	67	
	60	Aug. 22	13	120	
	57	Aug. 22	29	112	
	58	Aug. 22	130	1300	0.020
	56	Aug. 22	11	135	
	59	Aug. 22	16	125	
	55	Aug. 22	8.4	40	
	52	Aug. 22	72	830	
	61	Aug. 22	52	340	
	Median		16	135	

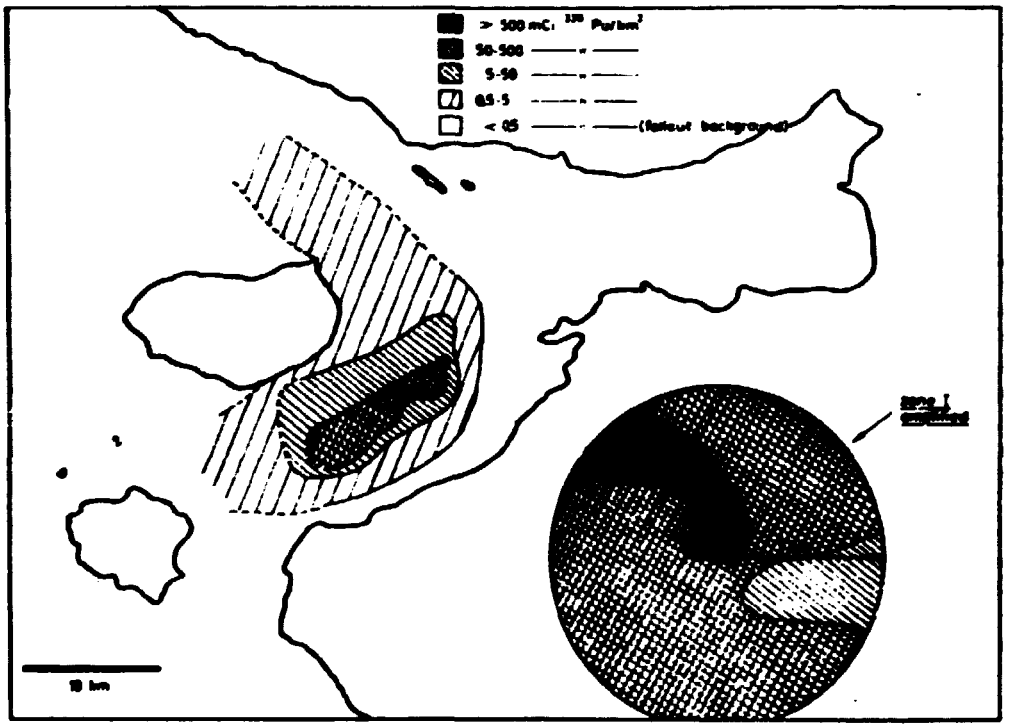

FIG. 3. The ^{239}Pu levels in August 1968 at the sea floor at Thule, Greenland.

this area is approx. 0.1 Ci/km², and the area is about 30 km². Hence the total deposition of plutonium within this area is 3 Ci. The fall-out background is estimated from location 12 to equal 0.2 mCi/km² or 0.1 pCi/g seabottom sediment, i.e. the same concentration as found by PILLAI[10] at the Californian coast.

It is evident that the measurements of the bottom sediments could not account for the total amount of plutonium (~25 Ci) supposed to have been released to the marine environment of Thule. As the highest level (1.3 Ci/km²) was found 1 km northwest of the point of impact, it seems probable that zone I with a total deposition of 0.4 Ci does not necessarily represent the maximum deposition in Bylot Sound. There may have been an area north of zone I, which might have shown even higher levels. One more thing should be kept in mind, namely that some plutonium might have attached itself to pieces of debris not included in the samples.

Bivalves

Table 7 shows that the soft tissue of bivalves collected in Bylot Sound in nearly all cases showed levels above the fall-out background, which is estimated at less than 10 pCi ^{239}Pu/kg soft tissue. The lowest levels were found at locations 26 and 2. Bivalves collected in October 1968 in Danish Inner waters contained 0.048 pCi/g ash or 7 pCi ^{239}Pu/kg fresh weight.

The plutonium activity in the bivalves was very unevenly distributed, double determinations (see stations 15, 17 and 32) showed a standard error far greater than the analytical error, which in most cases was less than 10%. The bivalves could thus not be considered in equilibrium with the sea water, and a calculation of a concentration factor would be meaningless. Besides, bivalves are always contaminated by the sediments, and it seems possible to calculate real concentration factors only when the bivalves have been free from contact with insoluble particulate activity.

Figure 4 shows the distribution of plutonium in bivalves collected in Bylot Sound. If we estimate the total amount of bottom animals at 450 g/m², and if all bottom animals were bivalves, the total integrated level of plutonium in bottom animals within the sampling area is 0.15 Ci, which is 5% of the estimated deposition in the sea bottom sediments (cf. above).

An analysis of variance of the results in Table 7 showed that *macoma* contained significantly higher levels than the other bivalves (approx. thirty times as much); whether this is due to the feeding habits of this species (*macoma* is a detritus eater) or to a greater superficial contamination of the animals is not clear. However, *macoma* is a small bivalve, and the contamination with bottom material might have been relatively great. It is especially remarkable that *macoma* from location 43, which is approx. 15 km northwest of the point of impact, showed plutonium levels nearly two orders of magnitude above the fall-out background, while the other species from this location differed only little from the background.

Bivalves have in the present study shown themselves to be suited for the environmental monitoring of plutonium in a marine environment.

Crustacea

In the trawl hauls performed around zone I (cf. Fig. 1) crustacea (mostly shrimps) were caught in appreciable quantities. Table 8 shows the plutonium levels in shrimps collected at Thule and Table 9 the levels in shrimps contaminated only with the fall-out plutonium. The median level in total shrimps collected at Thule was three orders of magnitude above the fall-out background. As in the case of bivalves it was evident that the activity was very unevenly distributed within the samples owing to the fact that the plutonium had been present as particulate matter rather than as a solution.

The fall-out background levels in bivalves and shrimps indicate that the concentration factor from sea water to soft tissue is greater for bivalves than for shrimps, namely ~3500 against 750. These factors are, however, very uncertain owing to the great analytical error in the determination of these weak samples, and to the fact that as mentioned earlier, the contamination of these bottom animals might be particulate. PILLAI *et al.*[17,18] found concentration factors of approx. 250–500 for mussels and of 2600 for mixed zooplankton.

Table 7. ^{239}Pu levels in bivalves collected in August 1968 at Thule, Greenland

Location (cf. Fig. 1)	Sample depth (m)	Sampling date	pCi/g ash from soft tissue	pCi/kg soft tissue	$^{239}Pu/^{239}Pu$	Species
Zone II						
26	94	Aug. 9	0.16	5.6B		*Musculus lenigatus and Astarte montagui*
16	94	Aug. 14	3.3	170		*Clinocardium ciliatum*
16	94	Aug. 14	2.6	107		*Hiatella striata*
16	94	Aug. 14	27	1000		*Mesodesma calcaria*
17	172	Aug. 7 & 10	100 ± 100	3000 ± 3000	0.020	*Clinocardium ciliatum & Mesodesma calcaria*
30	250	Aug. 16	51	2300	0.014	*Mesodesma calcaria*
29	97	Aug. 11	0.23	10		*Chlamys islandica*
29	97	Aug. 11	0.90	64		*Clinocardium ciliatum*
29	97	Aug. 11	0.20	6.7		*Hiatella striata*
29	97	Aug. 11	29	1500	0.013	*Mesodesma calcaria*
29	97	Aug. 11	0.53	22		*Musculus lenigatus, Astarte borealis & Astarte montagui*
27	30	Aug. 10	0.30	11		*Chlamys islandica*
27	30	Aug. 10	0.32	10		*Astarte montagui*
18	70	Aug. 14	0.43	22		*Serripes groenlandica & Clinocardium ciliatum*
18	70	Aug. 14	36	1700		*Astarte montagui, Mesodesma deauria & Hiatella striata*
34	140	Aug. 15	19	915	0.022	Various species, *Chlamys islandica, mesodesma, hiatella, clinocardium*
20	22	Aug. 14	0.67	18		*Chlamys islandica*
20	22	Aug. 14	0.22	5.9		*Musculus lenigatus & Astarte montagui*
19	155	Aug. 13	0.75	40		*Clinocardium ciliatum*
19	155	Aug. 13	8.4	450	0.023	*Mesodesma calcaria*
32	195	Aug. 13	98 ± 45	5400 ± 2400	0.022	*Mesodesma calcaria*
15	105	Aug. 13	16 ± 9	770 ± 430		*Mesodesma calcaria*
14	210	Aug. 13	12	640		*Musculus niger & Mesodesma calcaria*
12	90	Aug. 12	1.15	110		*Chlamys islandica & Clinocardium ciliatum*
12	90	Aug. 12	1.44	96	0.026	*Mesodesma calcaria & Hiatella striata*
12	90	Aug. 12	0.27	13		*Serripes groenlandicus & Astarte borealis*
2	1	July 25	0.23	4.7		*Mytilus edulis*
2	1	July 27	0.36	16		*Mytilus edulis*
43	100	Aug. 20	0.50	32		*Chlamys islandica*
43	100	Aug. 20	0.26	9.5		*Clinocardium ciliatum*
43	100	Aug. 20	0.55	21		*Musculus niger*
43	100	Aug. 20	0.35	17A		*Hiatella striata*
43	100	Aug. 20	10.5	500		*Mesodesma calcaria*
Median			0.75	40		

Table 7. (cont.)

Location (cf. Fig. 1)	Sample depth (m)	Sampling data	pCi/g ash from soft tissue	pCi/kg soft tissue	$^{238}Pu/^{239}Pu$	Species
Zone I						
52	188	Aug. 22	15	820		
53	203	Aug. 22	7.1	340		
54	217	Aug. 22	5.2	960	0.032	
55	178	Aug. 22	220	21000	0.013	
56	178	Aug. 22	12	720		Mixed samples, mostly of
57	185	Aug. 22	1000	76000	0.012	*macoma*, *Musculus niger*,
58	188	Aug. 22	140	8600	0.016	*hiatella*, & *astarte*
59	196	Aug. 22	32	8300		
60	198	Aug. 22	250	25000	0.015	
61	215	Aug. 22	53	7700	0.019	
Median			42	8000		

The shells of crustacea normally contained more ^{239}Pu than did the flesh, probably owing to surface contamination of the shells as the shrimps are bottom animals and thus live in close contact with the contaminated sea floor. It might, however, also be due to an accumulation of plutonium in the shells. E. E. WARD[11] has found that the concentration factor for lobster shells is two orders of magnitude as high as that for lobster flesh.

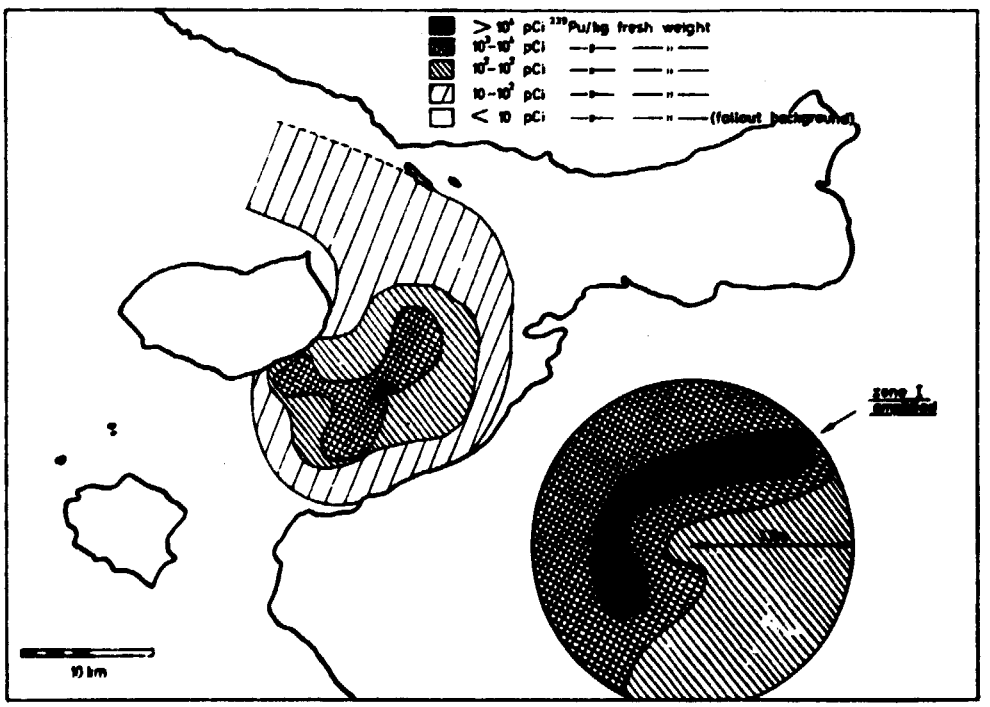

FIG. 4. The ^{239}Pu levels in bivalves collected in August 1968 at Thule, Greenland.

Table 8. ^{239}Pu levels in crustacea (shrimps) collected in August 1968 at Thule, Greenland

Location (cf. Fig. 1)	Sample depth (m)	Sampling date	pCi/g ash	pCi/kg fresh weight	$^{238}Pu/^{239}Pu$	Sample type
42	235	Aug. 18	0.55 ± 0.17	22 ± 6		Total animal
44	190	Aug. 20	5.3	140	0.028	Flesh
44	190	Aug. 20	35	2500	0.018	Shell
44	190	Aug. 20	38 ± 20	1900 ± 1000		Total animal
44	185	Aug. 20	45	1500	0.016	Flesh
44	185	Aug. 20	130	11000	0.013	Shell
44	185	Aug. 20	155	12000	0.017	Total animal
45	240	Aug. 20	3.5	95		Flesh
45	240	Aug. 20	3.9	280	0.033	Shell
45	240	Aug. 20	0.28	6.7A		Flesh
45	240	Aug. 20	1.7 ± 0.8	102 ± 50		Shell
47	160	Aug. 21	0.23	7.4		Flesh
47	160	Aug. 21	4.1	330		Shell
47	160	Aug. 21	43	3300		Total animal
59	196	Aug. 22	0.87	41		Total animal
Median			3.5	95		Flesh
Median			4.1	330		Shell
Median			38	1900		Total animal

Table 9. ^{239}Pu levels in shrimps collected outside the Thule area

Location	Sampling date	pCi/g ash	pCi/kg fresh weight
SW Greenland st. 3964	Spring 68	0.09 ± 0.03	3.8 ± 1.2B
Jacobshavn, Greenland	Summer 67	0.03	1.1A
Inner Danish waters st. 1459	19 Oct. 68	0.03	1.5B
Inner Danish waters st. 14588	18 Oct. 68	0.02	0.9A
Inner Danish waters st. 14589			
st. 14595	22 Oct. 68	0.04	2.2A
Median		0.03	1.5

Zooplankton

Figure 1 shows the location of the plankton hauls, and Table 10 gives the results. The ^{239}Pu levels in zooplankton are strikingly low compared with those in crustacea. The reason might be that the zooplankton had not been in contact with the bottom as had the shrimps. If the concentration factor found by PILLAI *et al.*[7] for zooplankton is valid for the Thule area, the expected level in zooplankton due to fall-out would be $2.10^{-3} \cdot 2600 = 5.2$ pCi ^{239}Pu/kg, which is close to the median level actually found. It has thus not been proved that the zooplankton samples contained any plutonium which could not be ascribed to fall-out.

Seaweed

Samples of sea plants (*Fucus* and *Laminaria*) were collected along the shores of Bylot Sound and Wolstenholme Fjord. Tables 11 and 12 show that the levels of plants collected around the accident area were not significantly different from the ^{239}Pu levels in brown algae collected at other locations in Greenland. The concentration factor in brown algae at Thule was estimated at $16/0.005 \sim 3.10^3$, which is twice as high as the factor of 1600 found for green algae (*Enteromorpha* spp) by PILLAI *et al.*[7]

Brown algae (*Fucus* and *Laminaria*) seem to be sensitive indicators of ^{239}Pu contamination of sea water.

Table 10. ^{239}Pu *levels in zooplankton collected in July–August* 1968 *at Thule, Greenland*

Location (cf. Fig. 1)	Sample depth (m)	Sampling date	pCi/g ash	pCi/kg fresh weight	Sample type
2	Tidal zone	July 25	0.41	30	*Gammarus*
37	Tidal zone	Aug. 16	0.48 ± 0.12	35 ± 9A	*Gammarus*
39	100–250	Aug. 17	0.18 ± 0.09	4.8 ± 2.4	Mixed zooplankton
38	40–200	Aug. 17	0.041 ± 0.009	0.90 ± 0.2	Mixed zooplankton
41	100–250	Aug. 18	0.12	3.2	Mixed zooplankton
Median			0.18	4.8	

Table 11. ^{239}Pu *levels in sea plants collected in July–Sept.* 1968 *at Thule, Greenland*

Location (cf. Fig. 1)	Species	Sampling date	pCi/g ash	pCi/kg wet weight
2	*Fucus sp.*	July 25	0.70 ± 0.05	35 ± 3
37	*Fucus sp.*	Aug. 9 Aug. 16	0.44 ± 0.09	17 ± 4
E	*Laminaria sp.*	Aug. 10	1.49	74
E	*Fucus sp.*	Sept. 18	0.28	15
G	*Laminaria sp.*	Sept. 17	0.16	5.9
G	*Fucus sp.*	Sept. 17	0.35	16
H	*Fucus sp.*	Sept. 18	0.33	15
Median			0.35	16

Table 12. ^{239}Pu *levels in sea plants collected in Greenland outside the Thule area*

Location	Species	Sampling date	pCi/g ash	pCi/kg fresh weight
Qanaq	*Fucus sp.*	Aug. 24, 68	0.32	15
Prins Chr. Sund	*Fucus sp.*	Summer 68	0.23	—
Danmarkshavn	*Fucus sp.*	Summer 68	0.79	—
Godthåb	*Fucus sp.*	Summer 67	0.21 ± 0.03A	—

Various bottom animals

The bottom samples collected by means of a 0.1 m^2 Petersen sampler in zone I contained various bottom animals, *polychaeta*, *echinodermata* and bivalves. These animals were separated from the bottom material and analysed for plutonium. Table 13 shows that the levels corresponded to the levels found in bivalves from zone I. This is not surprising, first because these animals live in close contact with the contaminated bottom sediments, secondly because they feed on each other; starfish e.g. eat bivalves.

Fish

A number of sea scorpions were jigged in the shallow waters near the coasts. Most of these samples showed no or very low levels of plutonium. Samples of polar cod caught in trawl hauls around zone I showed measurable, although not high, levels of plutonium. The highest level (470 pCi/kg) was found in a bottom fish, the Greenland halibut, (cf. Table 14), which is understandable as this species feeds on shrimps among other things. The median level in fish was 36 pCi/kg and could not be considered in equilibrium with the sea water and thus

Table 13. ^{239}Pu *levels in various bottom animals collected on* 22 *Aug. in zone I at Thule, Greenland*

Location (cf. Fig. 1)	Species	pCi/g ash	pCi/kg fresh weight	$^{239}Pu/^{239}Pu$
9 stations in zone I	Worm (*Polychaeta*)	230	30,000	0.011
60, 52	Starfish (*Asteroidea sp.*)	4.3	190	
53, 55, 59	Starfish (*Asteroidea sp.*)	0.39	190	
55	Starfish (*Asteroidea sp.*)	7.3	1120	0.024
59	Starfish (*Asteroidea sp.*)	2.2	490	
52	Corals	170	28,000	0.019

Table 14. ^{239}Pu *levels in fish collected in August* 1968 *at Thule, Greenland*

Location (cf. Fig. 1)	Species	Sampling date	pCi/g ash	pCi/kg fresh weight	$^{239}Pu/^{239}Pu$
21	Sea scorpion, roe	Aug. 7	0.023	0.26A	
21	Sea scorpion, bone	Aug. 7	0.012	1.32B	
40	Sea scorpion, total	Aug. 15	0.030	0.87A	
21	Sea scorpion, total	Aug. 7	0.68	14	
45	Polar cod	Aug. 20	0.41	19A	
47	Polar cod	Aug. 21	0.69	34	
44	Polar cod	Aug. 20	7.4	230	
44	Greenland halibut	Aug. 20	18	470	0.015
44	*Liparidae* and *Lycodidae* species	Aug. 20	5.6	260	
45		Aug. 20	0.47	35	
47		Aug. 21	0.83	40	
47		Aug. 21	0.58	38	
	Median of total fish		0.7	36	

could not be used for the calculation of concentration factors.

Sea birds

The median level of plutonium in entrails and intestinal contents of seabirds was 3.2 pCi/kg; this was close to the mean level in zooplankton, which is an important constituent of their diet. Samples of excrements from eiders contained relatively high plutonium levels. However, these samples were scraped from stone surfaces and thus inevitably contaminated with dust which might have contained plutonium.

A similar drawback attached to two samples of eiderdown collected from eiders' nests on the Manson and the Eiderduck islands. These samples contained respectively 84 and 184 pCi ^{239}Pu/kg. Eiderdown is of special interest because the Eskimos collect the down and clean them by a process, which is rather dusty and might thus constitute a risk for inhalation of contaminants adhering to the down.

Seals and walruses

Most of the seal and walrus samples were obtained from the Eskimos at Maniumak and Narssarssuk. Two seals were shot by members of the expedition. The plutonium levels were low, as shown in Table 16, in fact lower than in any other animal group examined. As regards the walruses this is not surprising, as these samples were from animals killed before the ice had melted in the contaminated area, and thus before the bivalves and other bottom animals had been contaminated.

Seal and walrus meat was not examined, considering the low intestinal and stomach levels.

Human urine

Urine samples from the Eskimos at Narssarssuk were collected three times; just after

Table 15. ^{239}Pu *levels in sea birds collected in August* 1968 *at Thule, Greenland*

Location (cf. Fig. 1)	Species	Sample type	Sampling date	pCi/g ash	pCi/kg fresh weight	Remarks
D	Eider	Excrements	Aug. 9	0.45	280	Sample contaminated with soil and dust
46	Eider	Excrements	Aug. 20	2.5	1550	Sample contaminated with soil and dust
E	Eider	Intestinal contents	Aug. 10	0.050	7	
40	Eider	Entrails	Aug. 18	0.040	2.3A	
40	Brunnich's guillemot	Stomach	Aug. 17	0.02	0.23B	
Zone I	Brunnich's guillemot	Entrails	Aug. 22	0.17	4.3A	
46	Black guillemot	Entrails	Aug. 20	0.15	3.2	
Median of entrail, stomach and intestinal content				0.05	3.2	

Table 16. ^{239}Pu *levels in seals and walruses collected in* 1968 *at Thule, Greenland*

Location (cf. Fig. 1)	Species	Sample type	Sampling date	pCi/g ash	pCi/kg fresh weight
C	Walrus	Stomach contents (fish)	April–May	0.029	0.25A
C	Walrus	Entrails	June–July	0.14	1.80
C	Walrus	Intestine	June–July	0.054	1.30
C	Walrus	Intestine	June–July	0.032	0.62
C	Walrus	Rectum	June–July	0.047	1.37
15	Harp seal	Intestinal contents	Aug. 8	0.057	1.00A
40	Ringed seal	Stomach contents	Aug. 17	0.19 ± 0.05	4.4 ± 1.2A
37	Harp seal	Stomach contents	Aug. 17	0.16	2.3
37	Harp seal	Stomach contents	Aug. 17	0.01	0.1B
37	Harp seal	Stomach contents	Aug. 17	0.037	0.49
Median				0.05	1.3

the accident in January, in September and in February 1969. A few of the samples from the first two collections showed measurable plutonium levels; however, the possibility that these samples had been contaminated during the sampling could not be excluded. Thus a new set of samples was collected in February, and none of these samples showed any traces of ^{239}Pu.

Plutonium-238

In the stronger samples (bottom sediments, bivalves and crustacea) it was possible to determine the activity ratio ^{238}Pu/^{239}Pu (cf. the tables). The ratio was determined in 26 samples, and the mean was 0.019 (1 SD = 0.006, 1 SE = 0.001). In fall-out from nuclear tests the ratio has varied through the years. Feely *et al.*[15] found that until 1964 high-altitude air

samples showed a fairly constant ratio averaging about 0.03. In precipitation collected in 1958–66 MIYAKE *et al.*[12] found a ratio of 0.039. For the period April 1963 to March 1965 BORTOLI *et al.*[10] found 0.019, (SD:0.003) in ground-level air, but in June–July 1966 the ratio had increased to 0.07 owing to the SNAP-9A burn-up, and in January 1968 the mean ratio in air in the northern hemisphere apart from Thule was 0.54 (SD:0.23).[11] In a brown alga[10] collected at the coast of California in 1964 the ratio was 0.03, i.e. close to the fall-out ratio.

^{239}Pu in fall-out derives partly from neutron capture of ^{238}U and partly from unfissioned, fissile material from the weapons. ^{239}Pu *is*, according to MIYAKE *et al.*,[12] produced in the bombs as a daughter product of ^{239}Np or ^{242}Cm, but might apparently also have come from unfissioned plutonium.

CONCLUSION

The B-52 accident at Thule in January 1968 raised the plutonium levels in the marine environment of Bylot Sound. The plutonium from the accident was mainly present as insoluble plutonium oxide particles. As in most cases the animals were contaminated with these particles, it was not possible to determine concentration factors from sea water to animals. The concentration in *sea water* increased by a factor of two. In *sea bottom sediments* the increase was one order of magnitude, beneath the impact area two orders of magnitude. *Bivalves* showed increased levels up to a distance of fifteen kilometres from the crash area. The median level in bivalves was one order of magnitude above the fall-out background, and in the crash area it was three orders above. *Macoma* contained more plutonium than the other species of bivalves. *Crustacea, polychaeta* and *echinodermata* showed median levels about 1000 times the fall-out background. *Fish* living near the sea bottom, e.g. the Greenland halibut, contained plutonium levels one to two orders of magnitude above the fall-out background. The other samples: *Zooplankton, sea plants, sea birds, seals* and *walruses* did not contain plutonium levels significantly different from the fall-out background. Plutonium was not detected with certainty in *human urine* samples.

The samples with significant ^{239}Pu levels from the accident contained 1.9% ^{238}Pu (as compared with ^{239}Pu). This percentage is close to that found in nuclear weapon fall-out prior to the SNAP-9A incident in 1964, and it is concluded that ^{238}Pu in fall-out may at least to some degree come from unfissioned plutonium in the weapons.

From ICRP's recommendations[13] for drinking water it is calculated that the maximum permissible daily intake of soluble plutonium for occupational exposure is 0.1 μCi. If, for instance, an Eskimo eats 100 g bivalves daily, which undoubtedly is an upper estimate of his bivalve consumption, the MPC in bivalves becomes 1 μCi ^{239}Pu/kg. Even the strongest sample of bivalves contained less than one tenth of this pessimistically estimated MPC value; furthermore most of the plutonium in bivalves was present as insoluble plutonium oxide.

As far as eiderdown is concerned, the annual permissible intake of insoluble ^{239}Pu from respiration is estimated at 73 nCi from ICRP.[13] The maximum concentration in eiderdown was 130 pCi/kg. Hence it seems unlikely that any Eskimo occupied with the cleaning of down might reach the permissible intake of ^{239}Pu into the lungs.

It is concluded that no samples showed plutonium levels that are supposed to be hazardous to man or higher animals in the Thule district or in any other part of Greenland.

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collection of snow samples from the 25 telestations in Greenland. Invaluable support was obtained from the American and Danish authorities at Thule during the whole period.

REFERENCES

1. E. P. Hardy, Jr. and Joseph Rivera, *USAEC report*, HASL-210 Appendix, 1 July (1969).
2. Y. Miyake, Y. Katsuragi and Y. Sugimura, Papers in Meteorology and Geophysics **19**, pp. 267 (1968).
3. M. Bortoli, P. Gaglione, A. Malvicini and E. van der Stricht, *Proc. of the 1st International Congress of Radiation Protection*, Rome 5–10 Sept. 1966 pp. 1053 Pergamon Press, Oxford (1968).

4a. R. S. Cambray, E. M. R. Fisher, G. S. Spicer, C. G. Wallace, and T. J. Webber AERE-R 4392 (1963) and 4687 (1964).

b. R. S. Cambray, E. M. R. Fisher, W. L. Brooks, A. Hughes and G. S. Spicer, AERE-R-4997, (1965).

c. R. S. Cambray, E. M. R. Fisher, W. L. Brooks and D. H. Peirson, AERE-R-5260 (1966).

d. R. S. Cambray, E. M. R. Fisher, W. L. Brooks and D. H. Peirson, AERE-R-5899 (1968).

5. *Report of the United Nations Scientific Committee on the Effects of Atomic Radiation* United Nations, New York (1969).
6. Chr. Vibe, The marine mammals and the marine fauna in the Thule district (Northwest Greenland) with observations on ice conditions in 1939–41, *Meddr. Grønland* **150**, 116 (1950).
7. K. C. Pillai, R. C. Smith and T. R. Folsom, *Nature, Lond.* **203**, 568 (1964).
8. V. T. Bowen and T. T. Sugihara, *Cycling and Levels of Strontium-90, Cerium-144 and Promethium-147 in the Atlantic Ocean.* Radioecology. Proceedings of the 1st National Symposium on Radioecology, Fort Collins, Colorado 10–15 Sept., 1961 135–139. Reinhold, New York (1962).
9. W. H. Langham, *USAF Nuclear Safety* **65**, 36 (1970).
10. T. R. Folsom, TID-23476, 1 Nov. (1966).
11. E. E. Ward, *Nature, Lond.* **209**, 625 (1966).
12. H. W. Feely, P. E. Biscaye, B. Davidson, H. Seitz, AD 642279, 1 July (1966).
13. *Radiation Protection.* Recommendations of the International Commission on Radiological Protection. Report of Committee II on Permissible Dose for Internal Radiation. (*ICRP Publication 2*) p. 233 Pergamon Press, New York (1959).
14. E. K. Markussen, *Radiochemical Procedures for the Determination of Plutonium in Environmental Samples.* Risö-M-1242 (1970).

Health Physics Pergamon Press 1977. Vol. 32 (April), pp. 271–284. Printed in Northern Ireland

ENVIRONMENTAL BEHAVIOUR OF PLUTONIUM ACCIDENTALLY RELEASED AT THULE, GREENLAND

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Abstract—The environmental contamination resulting from the B-52 accident in 1968 at Thule was studied by scientific expeditions in 1968, 1970 and 1974. The contamination was mainly confined to the marine environment, where plutonium was preferentially located in the sediments and the benthic fauna. Plutonium concentrations down through the sediment layers decayed exponentially with a half-depth of 1–2 cm. The horizontal distribution of the plutonium may be described by an exponential expression: $mCi\ ^{239,240}Pu\ km^{-2} = 460e^{-2.5R}$ or by a power function: $mCi\ ^{239,240}Pu\ km^{-2} = 370\ R^{-1.2}$, where R is the distance in km from the point of impact. The inventory of $^{239,240}Pu$ in the marine environment from the accident was estimated at 25–30 Ci. The amount of ^{238}Pu was ~0.5 Ci. The bottom animals, such as worms and molluscs, showed a horizontal distribution of radioactivity similar to that of the sediments. From 1968 to 1970 the $^{239,240}Pu$ concentrations in the biota decreased by an order of magnitude, since 1970 the decrease has been less evident. In 1970 and 1974 there were no indications of increased plutonium concentrations in surface seawater or in sea plants or zooplankton. Higher animals such as fish, seabirds and marine mammals have shown no tendency to increasing plutonium levels since the accident.

INTRODUCTION

Since the beginning of the Nuclear Age several cases of environmental plutonium contamination have been reported. The most widespread and well-studied contamination is the global fallout from nuclear weapons testing which has dissipated (325 ± 36) kCi plutonium in the biosphere (Har73). However, more concentrated contamination has been that from the reprocessing plant at Windscale, which has discharged approx $\sim 10^4$ Ci $^{239,240}Pu$ into the Irish Sea during the past 15 yr (He75). In comparison, the release from the B-52 accident at Thule Airbase, Greenland, in January 1968, when ~25 Ci were dissipated in the marine environment (Aa71a) is modest contamination. Although the Thule incident may thus seem trivial as regards the amount of plutonium involved, it is felt that the special circumstances such as the Arctic environment, the particulate nature of the plutonium, and the point source character of the release have made it worthwhile to monitor the environmental plutonium levels at Thule. The aims of the studies described herein have been to identify the area of distribution of the contamination and to elucidate a possible radioecological propagation of the plutonium.

The initial levels were established by the scientific expedition in 1968 (Aa71a); increased plutonium concentrations were measured as far as 15 km from the point of impact, and sediments and bottom animals in the impact zone showed plutonium levels 2–3 orders of magnitude above the Pu fallout background. The next scientific expedition took place in 1970. A summary of the results from that expedition (Aa71b) showed that the contamination had moved as far as 30 km from the point of impact; that the integrated plutonium level in the biomass of bottom animals had decreased by a factor of ~4; and that higher animals such as fish and seals did not show significantly higher levels than in 1968.

The third scientific expedition took place in August 1974; besides four Danish participants, Mr. David E. Robertson from Batelle Northwest Laboratories and Dr. Wayne C. Hanson from Los Alamos Scientific Laboratory were members of the party. This paper reports the results from the marine sampling aspects of the 1974 expedition and some of

the unpublished data from the 1970 expedition. To elucidate the time trend in the plutonium levels, a short summary of some of the 1968 data is included for comparative purposes.

MATERIAL AND METHODS

The environmental characteristics of the Thule locality have been described earlier (Aa71a) and are not repeated here. As previously the marine sampling was carried out by the Greenlandic ship "Aglanta." In comparison with 1970 and 1968, we extended the sampling in 1974 further away from the point of impact, the most distant sampling location being at a distance of 45 km (Fig. 1).

The 1974 expedition emphasized the collection of bottom sediments and bottom dwelling animals because sampling in 1968 and 1970 had shown that the highest plutonium levels were encountered in those types of samples. Two kinds of sediment samplers were used: a surface sampler, the PK sampler; and a bottom corer, the HAPS sampler that was developed by the Marine Biological Laboratory at Elsinore (Ka73). The PK sampler was also used in the earlier expeditions; it scrapes the uppermost layer of the sea bottom to a depth of ~1 cm over an rectangular area of 0.1 m^2 (Aa70). Experience showed that the sample depth varied with the nature of the bottom sediments; in muddy material the sample depth exceeded 1 cm and was somewhat shallower on a sandy bottom. The HAPS is in principle a corer with a top valve and a core catcher supported by a frame; it yielded well-defined samples and increased the effective sampling depth. The coring tube of stainless steel has a sampling diameter of 135 mm corresponding to a sampling area of 143 cm^2. When the core catcher closes, it may carry off a little surface material which contaminates the bottom layer of the sample; to prevent this, the lowest 1–2 cm of the sample core was discarded. The remaining part of the core is pushed out of the sampling tube by a piston and divided into 3 cm thick sections. The HAPS corer operated satisfactorily in Thule sea depths of >200 m provided that the bottom material was not stony.

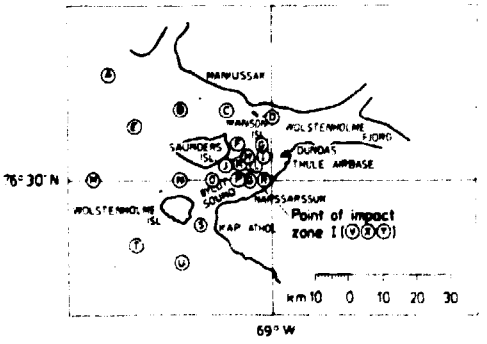

FIG. 1. The sample locations at Thule, Greenland (Location 19 is located in Bylot Sound at 76°27'8N, 69°37'5W).

Sediment samples were collected by both the PK-sampler and the HAPS corer at a number of locations. This comparison made it possible to empirically convert the surface data obtained by the PK-sampler to the total integrated level in the sediment column. The plutonium inventory in the uppermost cm of the sediments was estimated to contain one-third of the total plutonium content of the sediment layer.

As previously, the biological samples were kept frozen until processed in the laboratory. The radiochemical plutonium analysis followed the classical procedure (Ta71). The chemical yields were determined by ^{242}Pu or ^{236}Pu spikes supplied by Dr. John H. Harley, U.S.E.R.D.A. Health and Safety Laboratory, New York. The average yield of all sample types was 55%, and the minimum detectable activity for 4000 min counts was ~10 fCi of ^{239}Pu. The analyses of the samples that contained plutonium fallout background levels, were usually performed on 5–10 g ash, while the samples collected near the point of impact were normally analyzed in 0.5–1 g aliquots. Samples that contained <3 fCi ^{239}Pu per g of ash were generally below the detection limits of the procedures applied in this study.

To avoid cross-contamination, both sample collecting and laboratory processing were carried out on samples taken far from the point of impact before proceeding to those taken close to.

Due to the particulate nature of the Thule

contaminations the plutonium was very inhomogeneously distributed in the samples especially the sediment. Those samples were therefore analyzed as duplicates and in some cases even as triplicates.

The analytical errors, including counting error, were usually 10–20%, depending on the activity content. Samples with a counting error of more than 33% were denoted by ~0.

RESULTS AND DISCUSSION

Seawater

Only six 50-l. samples of unfiltered water were analyzed from the 1970 and 1974 samplings (Table 1) because 1968 plutonium levels in the Thule seawater was essentially that of the fallout background (~1 fCi ^{239,240}Pu l.$^{-1}$), apart from a few samples that probably contained particulates stirred up from the bottom sediments. In 1970, six samples collected off the west Greenland Coast from Godthåb (~64°N) to Thule (~76°N) showed a mean level of 1.6 ± 0.3 (S.E.) fCi ^{239,240}Pu l.$^{-1}$. Thus, neither in 1970 nor in 1974 did the Thule water contain levels significantly above fallout background, apart from the bottom sample from location G, sampled in 1974. Analysis of large-volume seawater samples collected in 1974 (Robertson, pers. comm.) showed that the surface water at Thule was typical of fallout background levels, whereas the near-bottom waters showed elevated plutonium levels of particulate nature due to resuspension of bottom sediments. Our samples of sea plants and zooplankton living in the surface water confirmed the fallout background concentrations of dissolved plutonium in the Thule water.

The low ^{239,240}Pu concentrations in Thule seawater were ascribed to the fact that the contamination at Thule consisted of highly insoluble plutonium oxide. Even in cases of releases of soluble plutonium to the aquatic environment, it has been general experience that the bulk of the plutonium is found in the sediments and not in the water masses. The Windscale studies (He75) have thus shown that only a small percentage of the total discharge of plutonium remains in the water, and similar results were obtained from studies in Lake Michigan of plutonium from fallout (Wa75), where ~96% of the fallout plutonium was transported to the sediment.

To what extent the plutonium in the Thule sediments dissolves has not yet been clarified, but leaching experiments and studies of interstitial water in the sediment cores (Robertson, pers. comm.) indicate some degree of dissolution.

Table 1. Plutonium in seawater from Bylot Sound collected in 1970 and 1974

Sample year	Location (Fig. 1)	Sample depth in m	fCi ^{239,240}Pu l.$^{-1}$
1970	C	112	0.6
1970	C	Surface	3
1970	V	Surface	2
1974	G	190	50
1974	P	240	1.5
1974	V	Surface	1.0

Detection limit 0.3 fCi l.$^{-1}$

Sea-bottom sediments

All but one of the sediment samples collected in 1968 and 1970, were surface sediments taken by the 0.1 m^2 PK-sampler. The exception was a 1970 core sample collected at the point of impact down to a depth of 10 cm by a Burke open-tube gravity corer, kindly placed at our disposal by Dr. V. T. Bowen of the Woods Hole Oceanographic Institute. This sample was divided into 3 sections of equal thickness. The analysis showed that ~$\frac{1}{3}$ of the total plutonium activity of the core were contained in the upper 3–4 cm. Assuming an exponential distribution of the plutonium concentration in the core, we estimated that the uppermost cm of the sediments contained ~$\frac{1}{4}$ of the total activity. Hence we could transform the results of the PK-samplings to total integrated plutonium in the sediment layers, under the assumption that the vertical distribution of Pu in all sediment samples collected both in 1968 and 1970 followed the pattern observed in the core sample collected at the point of impact in 1970. It was, of course, not satisfactory to base such sweeping conclusions on a single sample, and it was important to obtain a more comprehensive collection of core samples. This was made possible in 1974 by the HAPS sampler.

As shown in Fig. 2, the plutonium concentration in the sediment cores decreased exponentially with increasing sampling depth. The

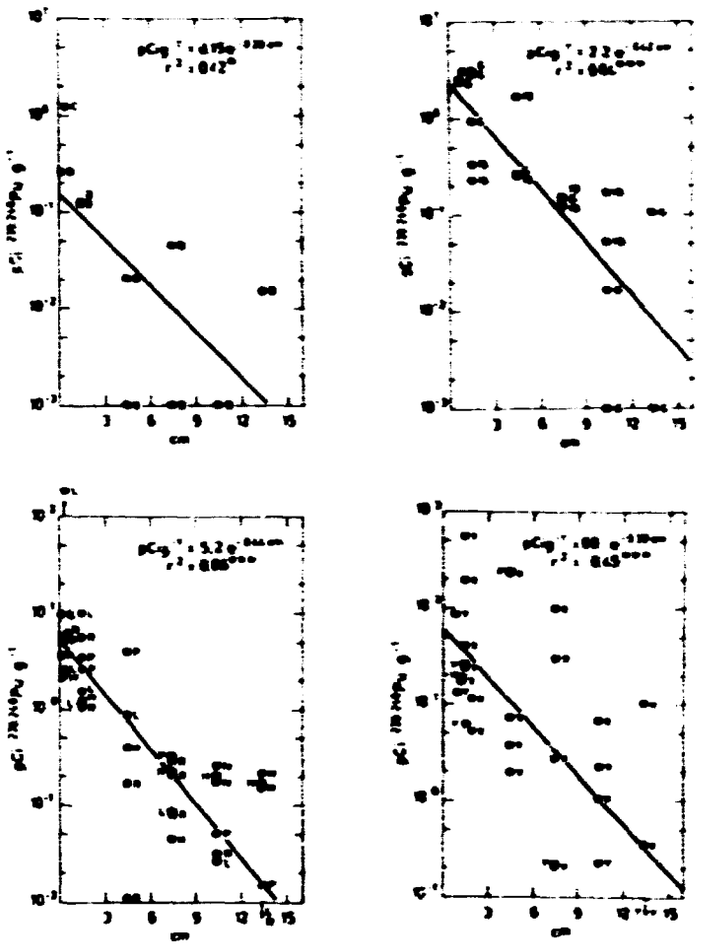

FIG. 2. The vertical distribution of $^{239,240}Pu$ in the sediments at various sampling locations at Thule. The abscissae are the depth in cm of the sediments; r^2 is the correlation coefficient between observed data and values calculated from the exponential equation (Significance levels: •: $P \leq 0.05$, • •: $P \leq 0.01$, • • •: $P \leq 0.001$)

four groups of sample locations, arranged according to decreasing distance from the point of impact, showed similar vertical distributions. Location H results were considered to be anomalously high, probably due to cross-contamination during sampling. The half-depth for the plutonium varied between 1.6 and 1.9 cm. The plutonium concentration C_{Rx} in the sediments R km from the point of impact and in the sample depth x cm may be described by:

$$C_{Rx} = K_R e^{-0.43x} \text{ pCi g}^{-1}, \quad (1)$$

where

$$K_R = 40e^{-0.31*R} \text{ pCi g}^{-1}, \quad (2)$$

as shown in Fig. 3.

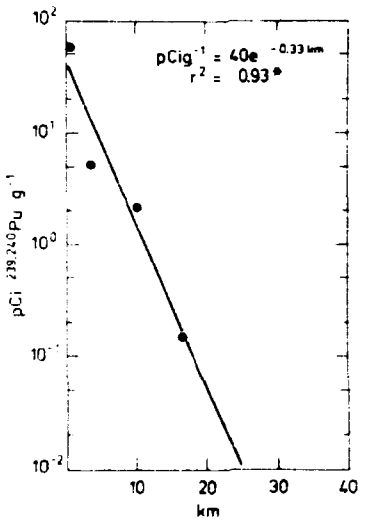

FIG. 3. The ^{239,240}Pu concentration in the sediment surface (0 cm depth) related to the distance (in km) from the point of impact

The Windscale studies (He75) have also demonstrated an exponential activity decrease down through the sediments. The half-depths in the Irish Sea were, however, considerably greater (4.6–8.6 cm) than in the present study, presumably because of the continuous nature of the releases from Windscale. In the studies of fallout plutonium in Lake Michigan (Ed75), plutonium was detectable only down to a depth of 6 cm, and there was no evident exponential decrease in plutonium concentrations down through the sediments; this was partly due to redistribution of previously deposited plutonium and partly to the dependence on the fallout rate.

The vertical distribution of the plutonium in sediments has received great attention and several explainations have been put forward. It has been proposed that the presence of plutonium in the deeper layers was due either to molecular diffusion and sorption/desorption taking place in the bulk sediment, or to biological reworking of the deposits, or to a combination of these processes (No72). The nearly constant relations between two so chemically different elements as Cs and Pu, which have been observed by several authors (No72; He75; Ed75), however, suggest, that physico-chemical mechanisms are less important.

In the Thule environment the strong tidal currents at the bottom stir up the sediments, as demonstrated by the particulate nature of plutonium found in the bottom water (Robertson, pers. comm.). This may result in both a horizontal and vertical dispersal of plutonium. Furthermore, we know that the worms contain plutonium in similar concentrations (per g of ash) as the upper sediment layers. Therefore we assume that these worms and other bottom animals may displace plutonium to greater depths in the sediments through their biological activities. Bowen (Bo75) suggested such a biological transport by fastmoving worms such as *Arenicola* and *Nereis*.

The inventory of plutonium in the Thule sediments may be estimated from equations (1) and (2). From the HAPS samples we calculated the average density in the upper 15 cm of the sediments at the 10 stations shown in Table 2 to be $(7.55 \pm 0.46) \cdot 10^9$ g cm^{-1} km^{-2}. There is no land area within 7.5 km from the point of impact and beyond 7.5 km land constitutes ~60% of the surface; thus, the inventory is calculated from integration of equations (1) and (2) to be: 25 Ci.

We may estimate the inventory more directly from the mCi km^{-2} data in Table 2, where the horizontal distribution of the plutonium deposit may be described by:

$$\text{mCi}\ ^{239,240}\text{Pu km}^{-2} = 460e^{-0.276R} \qquad (3)$$

where R is the distance in km from the point of impact. The correlation coefficient (r^2) between observed and calculated values was 0.60 and significant ($P \leq 0.01$). Analogous to the above calculation, we get: 29 Ci. The latter estimate is slightly greater because the density increases down through the sediments, which means that the deposition of plutonium (mCi km^{-2}) decreases more slowly with depth than the plutonium concentration (pCi g^{-1}).

Instead of the exponential expression (3) we may obtain a better description of the situation by a power model:

$$\text{mCi}\ ^{239,240}\text{Pu km}^{-2} = 370\, R^{-1.21}. \qquad (4)$$

Table 2. The vertical and horizontal distribution of plutonium at Thule in 1974. All values are in mCi ^{239,240}Pu km^{-2} in sediment sample layers

Location (Fig. 1)	Distance from point of impact (km)	HAPS sample section 0–3 cm	3–6 cm	6–9 cm	9–12 cm	12–15 cm	PK samples Sample depth in cm	PK samples Inventory down to sample depth	Integrated activity in total sediment layer calculated from HAPS and PK
C	17	—	—	—	—		0.8	4.8	16
D	16	3.0 3.1	~0 0.5	1.2 ~0	~0	0.4	0.2	0.4	2.5
G	9	17 11 34	5.4	3.0	0.4 ~0	2.4 ~0	1.2	11.6 12.1	30
H	3	25	11	1.5 7.7	7.2 4.9 5.6	5.3 6.5 4.9	0.6	9.7 165	97
K	1.4	—	—	—	—	—	1.1	3.3	118
L	4	18 156 26	16	1.6	0.5	~0	0.6	7.4 440	106
P	4	51	78	6.1	1.2	0.3	0.7	13 18	94
Q	3	—	—	—	—	—	0.3	4.8 13	44
R	4	107 49 20	4.0 0.2 16	2.0 4.9 8.7	0.8	~0	1.0	37	61
19	11	8.3 10.8	46 7.4	4.0 4.7	1.6 5.4	—	—	—	37
V	0	5800 260 270	75 39	4.3 57	25 54	8.5 1.4	2.4	165 153	2300
X	0.8	2600 540	4500 4400	2200 640	170	300	4.3	96 47	7400
Y	0.8	77	146	5.1	5.4	0.4	1.8	100 640	2300

Comments: ~0 means below detection limit (<0.1 mCi km^{-2})

The correlation coefficient (r^2) between observed and calculated values was 0.73 and highly significant ($P \leq 0.001$).The integrated plutonium level out to a distance of 45 km, where we measured levels not significantly different from the fallout background, becomes: 32 Ci power functions have also been used in inventory estimates by other investigators, such as around the Savannah River reprocessing plant (Mc75).

A comparison of the present estimates of the plutonium inventory in Thule with the previous ones of 17 Ci in 1968 and 24 Ci in 1970 (Aa71b) seems fairly satisfactory when we consider the great uncertainties of the earlier estimates because they were almost exclusively based on surface sediment samples. We conclude that the inventory of ^{239,240}Pu still present in the Thule sediments is 25–30 Ci, or ~400–500 g.

Seaweed

Laminaria and *Fucus* species were collected along the coasts of Bylot Sound in 1970 and 1974. The geometric mean levels were 2.1 pCi ^{239,240}Pu kg^{-1} wet weight (13 pCi kg^{-1} dry weight) in 1970 (7 samples) and 2.4 pCi ^{239,240}Pu kg^{-1} wet weight (16 pCi kg^{-1} dry weight in 1974 (3 samples) (cf. Table 3).

Concentration factors of the order of $2 \cdot 10^3$

Table 3. Plutonium in brown algae collected at Thule, 1970 and 1974

Sampling year	Locality (cf. Fig. 1)	Species	pCi ^{239,240}Pu kg^{-1} fresh weight	pCi ^{239,240}Pu kg^{-1} dry weight
1970	Narssarsuk	*Laminaria* sp.	—	6
1970	Saunders Island SW	*Fucus* sp.	—	24
1970	Wolstenholme Island N	*Laminaria* sp.	—	5
1970	Dundas	*Fucus* sp.	—	22
1970	5 km south of Manson Island	*Fucus* sp.	—	35
1970	Wolstenholme Island SE	*Fucus* sp.	—	9
1970	Manson Island	*Laminaria* sp.	—	12
1974	Narssarsuk	*Laminaria* sp.	0.42	3
1974	Dundas	*Fucus* sp.	8, 4	55, 25

were found for brown algae in Thule in 1968 (Aa71a). From this we may estimate the seawater level to be of the order of 1 fCi ^{239,240}Pu l.$^{-1}$, which is compatible with the observations in Table 1.

Zooplankton

Mixed samples of zooplankton were collected in Bylot Sound both in 1970 and 1974. The mean level of three samples in 1970 was 1.1±0.3 (S.E.) pCi ^{239,240}Pu kg^{-1} fresh weight; in 1974 we found 0.4 pCi ^{239,240}Pu kg^{-1} fresh weight.

In the plutonium studies in Lake Michigan (Wa75) it was observed that the concentration factor (CF) for plutonium followed the expression: CF = 200 (% ash)$^{1.4}$ for phytoplankton as well as zooplankton. In 1970 the mean ash content of the samples from Thule was 2.2% and in 1974 3.2%. If we assume that the Lake Michigan equation also applies to the marine environment at Thule, the expected concentration factors should be: $0.6 \cdot 10^3$ in 1970 and $1 \cdot 10^3$ in 1974 and the seawater levels: 1.8 fCi l.$^{-1}$ and 0.4 fCi l.$^{-1}$, respectively. Table 1 shows that these estimates are of the right order of magnitude.

Brittlestars (Ophiura) *and starfish* (Asterias)

Echinodermata (generally brittlestars) were present in most bottom samples and contained from 1 to nearly 6000 pCi ^{239,240}Pu kg^{-1} fresh weight. Although the highest plutonium concentrations were usually found closest to the point of impact, it was not possible to demonstrate any significant variation in the levels. Figure 4 shows a log–log plot of Pu concentrations in echinoderms found in 1970 and 1974 at various distances from the point of impact. The data indicates no time trend.

Molluscs

Bivalves were the most extensively sampled marine biota at Thule. Plutonium concentrations in their soft parts (Fig. 5) were related to the sampling distance from the point of impact and showed a marked decrease in the Pu values from 1968 to 1970, followed by a much slower decline to 1974 values. The decreased rate of change is presumably due to the dispersal and dilution with distance of the Pu. A

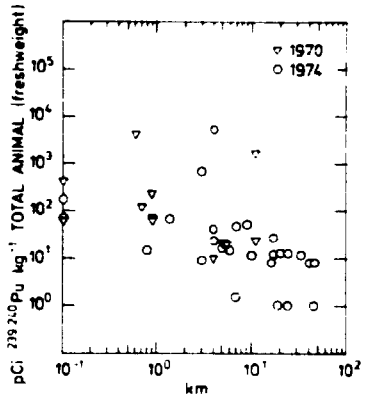

FIG. 4. The ^{239,240}Pu concentration in brittlestars and seastars collected at Thule in 1970 and 1974 related to the distance from the point of impact.

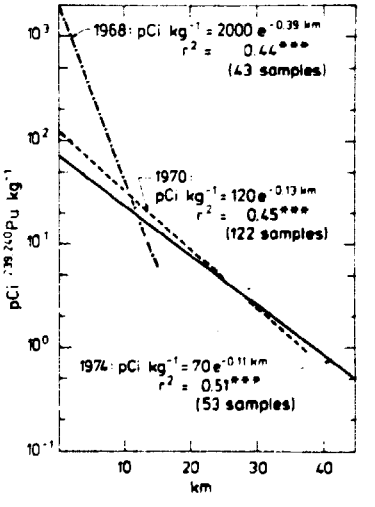

FIG. 5. The ^{239,240}Pu concentration in soft parts of bivalves collected in 1968, 1970 and 1974 as a function of distance from the point of impact; r^2 is the correlation coefficient between observed data and the values calculated from the exponential equations. (Significance level: * * *: 0,001, i.e. highly significant).

further dispersal from 1970 to 1974 is less evident.

The plutonium inventory in the bivalves was calculated in the same way as in the sediment, assuming that the weight of the soft parts of bivalves per m^2 is of the order of 100 g m^{-2} (Vi50).

1968: 0–7.5 km: 7 mCi; > 7.5 km: 0.7 mCi
Total 7.7 mCi
1970: 0–7.5 km: 1.1 mCi; > 7.5 km: 1.3 mCi
Total 2.4 mCi
1974: 0–7.5 km: 0.7 mCi; > 7.5 km: 1.2 mCi
Total 1.9 mCi

Again there was a rapid decrease from 1968 to 1970, followed by a more gradual decrease in plutonium concentrations.

Instead of the exponential curve shown for the 1974 data in Fig. 5 we propose a power model, as in the case of the sediments:

$$pCi\ ^{239,240}Pu\ kg^{-1} = 74.5\ R^{-0.89}.$$

The correlation coefficient (r^2) between observed and calculated values was 0.72 and highly significant ($p \leq 0.001$).

The integrated level in 1974 out to a distance of 45 km becomes: 1.4 mCi.

In the 1968 study (Aa71a) *Macoma* generally contained higher plutonium concentrations than the other species. An analysis of variance (ANOVA) of the data included in Table 4 shows significant variations between years (1970 > 1974), between sampling distances and between species (*Macoma*, *Hiatella* > *Leda*, *Clinocardium*, *Musculus*, *Chlamys*, *Astarte*, *Serripes*). The concentration of plutonium in *Macoma* and *Hiatella* was ~3 times greater than that in *Chlamys*, *Astarte*, and *Serripes*.

The ANOVA revealed no significant interactions between the three parameters, probably due to the large residual error.

Because it was impossible in laboratory processing to avoid some contamination of the mollusc soft parts by sediments, it was not possible to determine whether or not the plutonium in the animals had been metabolized. To clarify this point we analyzed 29 samples of shells that were first washed in dilute hydrochloric acid to remove any surface contamination from sediments. The results (Table 5) indicated that the levels in the shells followed the same pattern with distance from the point of impact as did the soft parts. The concentration in the shells was ~2.5 times higher than in the soft parts. Experimental observations (Fo75) have shown that shells of mussels contain 4–20 times higher concentrations than the soft parts. The lesser ratio in the present material may be due to the unavoidable contamination of the soft parts by the sediments, as mentioned above.

Plutonium concentrations in the soft parts of snails (*Buccinum*, Table 6) corresponded with good approximation to the geographical distribution of similar measurements in other molluscs, but the levels were generally lower in snails. The activity followed the equation:

$$pCi\ kg^{-1} = 16e^{-0.12\,R}.$$

If we compare this equation with that for molluscs in Fig. 5, we find the plutonium

Table 4. Plutonium (pCi ^{239,240}Pu kg^{-1}) in mollusc soft parts collected at Thule in 1970 and 1974 at various distances from the point of impact

Distance interval (km)	Year	*Chlamys islandica*	*Hiatella sinuata*	*Macoma calcarea*	*Clinocardium ciliatum*	*Musculus niger*	*Astarte montagui*	*Serripes groenlandica*	*Leda pernula*	Geom. mean	S.E. factor
>30–45	1970	2.1 (2)	3.5 (2)	1.0 (1)	1.0 (1)		2.0 (1)	2.0 1)		1.8	1.2
	1974	1.9 (1)		2.2 (3)	1.3 (2)	16.1 (1)				3.1	1.8
>20–30	1970	3.0 (3)	9.6 (3)	6.4 (5)	5.7 (3)	4.4 (3)	3.6 (4)	2.0 (3)		4.4	1.2
	1974		3.5 (1)	2.8 (2)	3.5 (2)					3.3	1.1
>15–20	1970	6.0 (1)	5.0 (1)	31 (2)		2.0 (1)	2.0 (1)			5.2	1.7
	1974		9.0 (1)	27 (2)	3.0 (1)		1.8 (1)	2.5 (1)		5.0	1.6
>10–15	1970	11.0 (1)	24 (2)	22 (3)	9.6 (2)	22 (1)	3.0 (1)			12.4	1.4
	1974										
>7.5–10	1970	3.8 (2)	27 (2)	32 (3)	19.6 (2)	35 (1)	20 (1)			18.9	1.4
	1974			26 (2)	15.5 (1)					20	1.3
>5–7.5	1970	5.7 (2)	49 (1)	85 (2)	8.0 (1)	11.3 (3)	30 (1)		44 (1)	22	1.5
	1974	12.8 (2)	25 (1)	17.4 (2)	13.2 (2)					16.5	1.2
>3–5	1970	30 (2)	60 (2)	52 (6)	54 (4)	12.8 (2)	17.2 (5)		70 (2)	36	1.3
	1974	10.1 (1)	83 (1)	18.2 (4)	14.0 (1)		3.9 (1)			15.3	1.6
>1–3	1970			636 (3)	26 (1)	440 (2)			290 (1)	220	2.1
	1974			260 (2)						260	1.0
>0–1	1970		670 (3)	3000 (5)	171 (4)	85 (3)			172 (3)	350	1.9
	1974	169 (2)	260 (2)	230 (4)	270 (1)					230	1.1

The figures are the geometric means of the number of samples shown in brackets.
1 kg fresh weight of soft parts ~45 g ash ~215 g dry matter.

Table 5. Plutonium in shells of molluscs collected in Thule 1974

Location (Fig. 1)	Distance from point of impact in km	Species	pCi ^{239,240}Pu g^{-1} ash
A	45	*Macoma calcarea*	0.01
		Clinocardium ciliatum	~0
B	24	*Serripes groenlandica*	0.05
		Clinocardium ciliatum	~0
C	17	*Macoma calcarea*	0.07
		Macoma calcarea	0.02
D	16	*Hiatella arctica*	0.02
		Astarte montagui	0.03
		Hiatella arctica	0.05
E	33	*Macoma calcarea*	0.01
		Musculus laevigatus	~0
		Clinocardium ciliatum	~0
		Macoma calcarea	0.06
F	7	*Chlamys islandica*	0.05
		Clinocardium ciliatum	0.03, 0.02
		Astarte montagui	0.04
G	9	*Macoma calcarea*	0.10
		Macoma calcarea	0.19
I	6	*Clinocardium ciliatum*	0.08
		Chlamys islandica	0.04
		Hiatella arctica	0.06
M	40	*Astarte montagui*	0.01
P	4	*Clinocardium ciliatum*	0.88
R	4	*Astarte montagui*	0.03
V	0	*Macoma calcarea*	0.59
		Clinocardium ciliatum	1.56
X	0.8	*Macoma calcarea*	0.48
Y	0.8	*Hiatella arctica*	0.22

Comment: ~0 means below detection limit (<0.005 pCi g^{-1} ash)

Table 6. Plutonium in soft parts of snails (Buccinum) collected at Thule, 1974

Location (Fig. 1)	Distance from point of impact in km	pCi ^{239,240}Pu kg^{-1} fresh weight
B	24	~0
E	33	~0, 5
F	7	2
I	6	6
N	19	0.4
O	10	5, 7
S	20	2
V	0	37

Comment: ~0 means below detection limit (<0.1 pCi kg^{-1}) 1 kg fresh weight ~37 g ash ~300 g dry matter

concentration in the soft parts of snails to be 4–5 times lower than that of bivalves.

Worms

As the plutonium contamination in the sediments approximately decreased exponentially with distance from the point of impact, it was obvious also to relate the plutonium levels in the bottom animals to the distance. Worms (Polychaeta) contain sediments in their digestive tracts, and a rather close relationship to the radioactivity concentrations in sediments was to be expected. Figure 6 shows a comparison. A three-way ANOVA of data of surface sediments and the worms from 1970 and 1974, grouped according to distance from the point of impact, showed no interaction between sample species and years, nor between years and locations or locations and species; neither was there any significant difference between sampling years. The plutonium concentration in the ashed worms was 2.8 times less than the concentration in sediments, and the values decreased exponentially with distance from the point of impact. It is calculated that, as regards the plutonium concentration in pCi g^{-1} of ashed material, a worm sample in the present material corresponds to a sediment sample collected down to a depth of 8.4 cm. According to equation (1) we may estimate the sample Pu content to be 96.5% of the total plutonium of the sediment column. From the ten HAPS cores collected we estimate the ash weight per m^2 of the 0–8.4 cm sediment layer to be 58.8 kg.

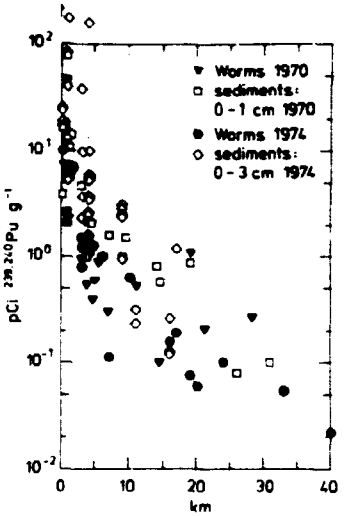

FIG. 6. The ^{239,240}Pu concentrations in ash of worms and surface sediments samples collected in 1970 and 1974 related to the distance from the point of impact.

The plutonium content of the combined worm material collected in 1970 and 1974 (Fig. 6) follows the expression:

$$pCi\,g^{-1} = 2.47 \cdot e^{-0.13\,R}$$

or

$$mCi\,km^{-2} = 151e^{-0.13\,R}$$

The integrated plutonium level becomes ~31 Ci which is in agreement with the estimates based on sediments.

We may conclude that worms, due to their ingestion of sediments, may be considered to

be "sediment equivalents" for the purpose of inventory determinations after an appropriate "calibration."

Shrimps (Arcturus)

Both in 1970 and in 1974 shrimps were collected from the point of impact out to a distance of ~40 km (Fig. 7). Similar to the echinoderms, there was a slight decrease in the plutonium levels with distance, but less definitive than in the case of worms and bivalves. The explanation may partly be the greater mobility of the crustaceans (and echinoderms), partly the more pelagic nature of these animals.

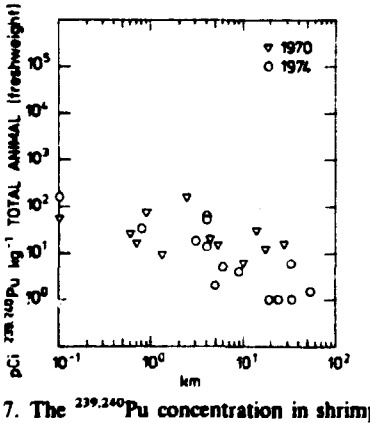

FIG. 7. The $^{239,240}Pu$ concentration in shrimps collected at Thule in 1970 and 1974 related to the distance from the point of impact.

In a few cases shells and flesh of shrimps were analyzed separately; the mean ratio between the plutonium concentrations in the two parts was 6; in 1968 the mean ratio of five samples was 15 (Aa71a). Experimental uptake studies (Fo75) have shown a concentration ratio between shells and soft parts of 3.3. The shells of shrimps from Thule have probably been contaminated by particulate plutonium from adhering sediments, which fact may have increased the ratios observed.

Fish

Eleven samples of fish (sea scorpion, Polar cod, *Agonida*, *Lycodida*) caught in Bylot Sound in 1970 and three samples from 1974 were analyzed for plutonium. The geometric

Table 7. Plutonium in fish samples caught in Bylot Sound in 1970 and 1974

Sampling year	Species	Sample type	pCi $^{239,240}Pu$ kg^{-1} fresh weight
1970	Polar cod	Total fish	2.5, 5
1970	Polar cod	Total fish	400
1970	*Lycodida*	Total fish	20
1970	*Lycodida*	Total fish	2
1970	*Lycodida*	Total fish	4.6
1970	*Lycodida*	Total fish	2
1970	*Agonida*	Total fish	5
1970	*Agonida*	Total fish	9
1970	*Agonida*	Total fish	4
1970	Sea scorpion	Flesh	0.2
1970	Sea scorpion	Liver	0.8
1974	Polar cod	Total fish	~0
1974	Sea scorpion	Total fish	10
1974	Sea scorpion	Liver	0.3
1974	Unidentified	Total fish	1.5

Comments: ~0 means below detection limit (<0.1 pCi kg^{-1})
Total fish: 1 kg fresh weight ~44 g ash ~220 g dry matter

mean levels of the two samplings (Table 7) were 4 and 1 pCi $^{239,240}Pu$ kg^{-1} fresh weight, respectively, being an order of magnitude less than values determined in 1968.

Birds

Only a few samples of seabirds (guillemots) were analyzed in 1970 and 1974. The results are shown in Table 8. As in the case of fish,

Table 8. Plutonium in samples of seabird, shot at Thule in 1970 and 1974

Sampling year	Species	Sample type	pCi $^{239,240}Pu$ kg^{-1} fresh weight
1970	Black guillemot	Entrails	3.5
1970	Brünnich's guillemot	Liver	128
1970	Brünnich's guillemot	Entrails	~0
1970	Brünnich's guillemot	Liver	~0
1970	Brünnich's guillemot	Entrails	~0
1974	Brünnich's guillemot	Entrails	0.3

Comments: ~0 means below detection limit (<0.1 pCi kg^{-1})

there was no evidence of any increase of plutonium concentrations since 1968.

Walrus and seal

The last link in the marine foodchain at Thule before man (and polar bear) is occupied by marine mammals. Table 9 shows that neither in 1970 nor in 1974 did the sampling (mostly entrails) of seal and walrus show any general indication of increased plutonium levels, as compared with the observations in 1968 (Aa71a). The geometric mean of all samples was 0.5 pCi $^{239,240}Pu$ kg^{-1} both in 1970 and 1974. The higher levels were mostly found in the undigested contents of intestine and stomach. We therefore conclude that no significant amount of plutonium from the Thule accident has been transferred to marine mammals.

Table 9. Plutonium in marine mammals shot at Thule in 1970 and 1974

Sampling year	Species	Sample type	pCi ^{239,240}Pu kg^{-1} fresh weight
1970	Ringed seal (*Phoca hispida*) I	Entrails	~0
1970	Ringed seal II	Entrails	1.1
1970	Ringed seal III	Entrails	0.1, 3.6
1970	Greenland seal (*Phoca groenlandica*)	Entrails	~0
1970	Seal I (unspecified)	Entrails	~0
	(unspecified)	Entrails	2.2
1970	Ringed seal II	Contents of intestine	0.8
1970	Ringed seal I	Liver	~0
1970	Ringed seal III	Liver	0.6
1970	Seal II (unspecified)	Liver	1.0
1970	Greenland seal	Liver	3.9
1974	Walrus I (*Odobenus rosmarus*)	Contents of intestine	3.3
1974	Walrus II	Contents of intestine	5.5, 1.6, 5.5
1974	Walrus I	Intestine	0.1
1974	Walrus II	Intestine	0.7
1974	Walrus II	Intestine	~0
1974	Walrus I	Stomach contents	1.6
1974	Walrus III	Stomach contents	1.4
1974	Walrus I	Stomach	0.2
1974	Walrus II	Stomach	0.04
1974	Walrus III	Stomach	0.8
1974	Walrus I	Liver	0.17
1974	Walrus II	Liver	0.20
1974	Walrus III	Liver	0.03

Comment: ~0 means below detection limit (<0.1 pCi kg^{-1})

The Eskimos occasionally collect bivalves from the walrus stomachs and thus have a direct access to the more contaminated links of the marine foodchain. The high degree of discrimination against plutonium by the gastrointestinal tract, however, reduces the importance of this type of exposure; this was also demonstrated by the low Pu levels found in walrus although they may have fed on contaminated bivalves.

Summary

The plutonium concentrations determined in environmental samples collected in 1970 and 1974 at Thule can be used for an estimate of the inventories in the various compartments of the benthic food-chain. The sediments contain practically all the plutonium released as a result of the accident in 1968. The benthos contains less than 1‰ of the total Pu-inventory (Table 10). Molluscs are estimated to contain the major part of the plutonium in the biota. The transfer-coefficient to the molluscs is an order of magnitude greater than the transfer-coefficient to brittlestars and shrimps. If, however, we compare the transfer-coefficients based on concentrations (Ci · g^{-1} · yr pr. Ci released), there is no significant difference between molluscs (soft parts), brittlestars and shrimps. We may thus

Table 10. Estimates of ^{239,240}Pu inventories and transfer-coefficients in the environment of Thule

Sample	Estimated biomass (Vi50) g m^{-2}	Inventory in Ci 1970	Inventory in Ci 1974	Transfer from the release of 1 Ci in Ci · yr
Molluscs:				
Soft parts	10^2	$2.4 \cdot 10^{-3}$	$1.9 \cdot 10^{-3}$	$1.5 \cdot 10^{-3}$
Shells	10^2	$6 \cdot 10^{-3}$	$5 \cdot 10^{-3}$	$4 \cdot 10^{-3}$
Brittlestars (total animal)	$3 \cdot 10$	$3 \cdot 10^{-3}$	$8 \cdot 10^{-4}$	$6 \cdot 10^{-4}$
Shrimps (total animal)	$2 \cdot 10$	1.10^{-3}	$2 \cdot 10^{-4}$	$2 \cdot 10^{-4}$
Worms (total animal)	10	$5 \cdot 10^{-4}$	$5 \cdot 10^{-4}$	—
Sediments	—	$3 \cdot 10$	$3 \cdot 10$	—

The inventories of the molluscs were taken from the text. Figures 4 and 7 were used for the estimates of brittlestars and shrimps. The inventory in worms was calculated from the exponential equation for worms.

expect nearly the same Pu concentrations in these animal groups from a given plutonium release to the sea. In Table 10 it has been assumed that the decay of the Pu-inventories observed from 1970 to 1974 would continue as single, exponential decays to infinity. The infinite integral of the respective exponential expressions calculated for the various sample types divided by 30 thus corresponds to the transfer-coefficient for the sample in question in the Thule environment (Ci · yr pr. Ci released).

The estimates are preliminary, and it is probably an over-simplification to expect the decay of the plutonium concentrations in the biota at Thule to follow a single, exponential decay. From 1968 to 1970 we observed a faster decay in molluscs than from 1970 to 1974, and we can expect the decay rates to decrease with time a factor, which will increase the transfer-coefficients for the various biota. Furthermore, it should be noted that the estimates of the biomass of the various animal groups in the Thule area only indicate the order of magnitude.

As regards the transfer of plutonium from the sediments to the seawater we may calculate K_d values [= (pCi g^{-1} surface sediments/pCi ml^{-1} sea water)] from the data given in Table 1 and Fig. 2. The geometric mean of these K_d values was ~10^6 ml g^{-1} (range: $4 \cdot 10^4 - 6 \cdot 10^7$).

In a study of ^{239,240}Pu in groundwater and groundwater particulates at Enewetak Atoll (No75) K_d values between 10^4 and 10^6 ml g^{-1} were observed. In a laboratory experiment with seawater and contaminated soil from

Enewetak Atoll a K_d mean value $8 \cdot 10^4$ ml g^{-1} was measured. The high K_d values at Thule were found near to the point of impact, where we expect plutonium particulates to have largest grain size. We therefore conclude that the plutonium from the Thule accident probably shows higher K_d values, and thus lesser solubility, than the fallout plutonium found at Enewetak Atoll.

Plutonium-238

In a number of the samples collected near the point of impact ^{238}Pu was determined together with ^{239,240}Pu. The mean percentage of ^{238}Pu as compared with ^{239,240}Pu was 1.9 ± 0.1 (SE), equal to the mean observed in 1968 (Aa71a). It has been suggested (Han75) that some physical separation process perhaps due to radioactive decay, may account for an apparently increased biological availability of ^{238}Pu. Although Table 11 shows a mean ratio in bivalves of 2.1, as compared to 1.7 in sediments and 1.8 in worms, the data were too few to prove that the ^{238}Pu/^{239,240}Pu ratio was significantly higher in bivalves than in sediments. Robertson (pers. comm.) has performed a more comprehensive study of ^{238}Pu in the Thule sediments and demonstrated a variation of the ^{238}Pu/^{239,240}Pu ratio with depth which may be a result of biological activity in the sediments.

Table 11. ^{238}Pu/^{239,240}Pu in sediments, bivalves and worms collected near the point of impact at Thule in 1974

Location (Fig. 1)	Sample type	^{238}Pu/^{239,240}Pu × 100
Q	PK-sediment (0–3 mm)	1.6
X	PK-sediment (0–4.3 cm)	1.7
Y	PK-sediment (0–1.8 cm)	1.9
V	PK-sediment (0–2.4 cm)	1.6
X	*Macoma calcarea* (soft parts)	2.7
Y	*Macoma calcarea* (soft parts)	2.4
V	*Macoma calcarea* (soft parts)	1.7
V	*Macoma calcarea* (soft parts)	1.8
Y	*Cingulina ciliatum* (soft parts)	1.7
Y	*Chlamys islandica* (soft parts)	2.3
V	*Mizuhopecten arctica* (soft parts)	2.2
V	*Mizuhopecten arctica* (soft parts)	1.8
V	Mixed sample of bivalves (soft parts)	1.8
X	Worms	2.1
Y	Worms	1.6
V	Worms	1.6
V	Worms	1.9

Table 12. Summary of ^{239,240}Pu data from the sample collections in 1968, 1970 and 1974

Sample	Unit		1968 I 0–1 km	1968 II >1 km	1970 I 0–1 km	1970 II >1 km	1974 I 0–1 km	1974 II >1 km
Seawater (surface (~100 m)	fCi l^{-1}	max.	12	67	3	—	—	—
		min.	4	2	2	—	—	—
		geometric mean	6	6	2	1	1	—
			(4)	(8)	(2)	(1)	(1)	
"PK" Sediments (0–1 cm) (ash)	pCi g^{-1}	max.	130	16	86	4.7	50	78
		min.	7	0.1	6	0.4	8	0.3
		geometric mean	23	1.0	13	1.2	17	5.0
			(9)	(7)	(9)	(10)	(3)	(9)
Sea plants (wet weight)	pCi kg^{-1}	max.	—	74	—	6	—	8
		min.	—	6	—	1	—	0.4
		geometric mean	—	19	—	2.1	—	2.4
				(7)		(7)		(3)
Worms (ash weight)	pCi g^{-1}	max.	—	—	46	1.1	10.5	6.9
		min.	—	—	0.3	0.1	2.1	0.06
		geometric mean	230	—	3.4	0.48	5.7	0.54
			(1)	—	(6)	(11)	(4)	(15)
Bivalves (soft parts) (fresh weight)	pCi kg^{-1}	max.	76,000	5,400	73,000	13,000	1900	300
		min.	320	5	50	1	75	2
		geometric mean	4,600	83	390	23	240	15
			(10)	(33)	(15)	(79)	(12)	(30)
Brittlestars Seastars (fresh weight)	pCi kg^{-1}	max.	1,120	—	4,400	1,700	250	64
		min.	190	—	10	10	62	1
		geometric mean	380	—	140	44	81	9
			(4)		(7)	(7)	(4)	(14)
Shrimps (fresh weight)	pCi kg^{-1}	max.	—	12,000	76	170	160	64
		min.	—	22	16	1	33	1
		geometric mean	41	1,130	35	16	72	9
			(1)	(4)	(2)	(10)	(2)	(8)
Fish (fresh weight)	pCi kg^{-1}	max.	—	470	—	400	—	10
		min.	—	1	—	0.2	—	<0.1
		geometric mean	—	40	—	4	—	1
				(10)		(11)		(4)
Birds (Fresh weight entrails)	pCi kg^{-1}	max.	—	7	—	130	—	—
		min.	—	0.2	—	<0.1	—	—
		geometric mean	—	2.2	—	0.9	—	0.3
				(5)		(9)		(1)
Seal and walrus (entrails, etc.) (fresh weight)	pCi kg^{-1}	max.	—	4.4	—	3.9	—	5.5
		min.	—	0.1	—	<0.1	—	<0.1
		geometric mean	—	1.0	—	0.5	—	0.5
				(10)		(12)		(15)

(figures in brackets indicate the number of samples).
I: *Zone* I, less than 1 km from the point of impact
II: *Zone* II, *outside zone* I

CONCLUSIONS

The plutonium contamination at Thule resulting from the B-52 accident in 1968 was mainly confined to the sediments which contained ~25–30 Ci. Beyond 40 km from the point of impact the environmental plutonium levels were not significantly above the fallout background.

The vertical distribution of the plutonium activity in the sediments decreased exponentially with a half-depth of ~1.7 cm. The total deposit in the sediments decreased exponentially with distance from the point of impact. The half-distance was estimated to be ~3 km. Marine worms and molluscs also showed exponentially decreasing plutonium concentrations with increasing distance, but the half-distance was 5–6 km, longer than for the sediments, probably a result of biological transport of the plutonium. The plutonium levels of other bottom animals, such as brittlestars and shrimps, also decreased with distance from the point of impact but less markedly than the worms and molluscs.

When compared with the initial sampling in 1968, the Pu concentrations in 1970 and 1974 were generally lower by an order of magnitude; the decrease from 1970 to 1974 was less marked. Molluscs, which constituted the largest number of samples, contained in 1974 ~60% of the plutonium concentration found in 1970, but for the other samples the decrease was not evident. The plutonium levels in sediment and worms, for example, did not change significantly from 1970 to 1974. We may, therefore expect a rather slow reduction in the plutonium levels of the marine environment at Thule from now on.

Neither in 1970 nor in 1974 did we find evidence of any significant increase of the plutonium concentration in the higher animals such as fish, seabirds and marine mammals at Thule. The plutonium originating from the accident has to date been confined to the bottom fauna, and man has not been at risk.

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REFERENCES

Aa71a Aarkrog A., 1971, "Radioecological Investigations of Plutonium in an Arctic Marine Environment," *Health Phys.* **20**, 31.

Aa71b Aarkrog A., 1971. "A Re-examination of Plutonium at Thule, Greenland, in 1970," *Proc. Symp. on Radioecology* p. 1213. Rome, Italy.

Aa70 Aarkrog A. 1970, "Radioecological Investigations," Project Crested Ice., Risö Report No. 213, p. 74.

Bo75 Bowen V. T., Livingston H. D. and Burke J. C., 1975 "Distributions of Transuranium Nuclides in Sediment and Biota of the North Atlantic Ocean," *Proc. Symp. on Transuranium Nuclides in the Environment* p. 107, San Francisco, U.S.A.

Ed75 Edgington D. N. and Robbins J. A., 1975, "The Behaviour of Plutonium and other Long-lived Radionuclides in Lake Michigan. II. Patterns of Deposition in the Sediments, " *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment*, p. 245, Otaniemi, Finland.

Fo75 Fowler S., Heyrand M. and Beasley T. M., 1975, "Experimental Studies on Plutonium Kinetics in Marine Biota," *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment*, p. 157, Otaniemi, Finland.

Han75 Hanson W. C., 1975, "Ecological Considerations of the Behaviour of Plutonium in the Environment," *Health Phys.* **28**, 529.

Har73 Hardy E. P., Krey P. W. and Volchok H. L., 1973, "Global Inventory and Distribution of Fallout Plutonium," *Nature, Lond.* **241**, 444.

He75 Hetherington J. A., Jefferies D. F. and Lovett M. B., 1975, "Some Investigations into the Behaviour of Plutonium in the Marine Environment," *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment*," p. 193, Otaniemi, Finland.

Ka73 Kanneworff E. and Nicolaisen W., 1973, "The HAPS a Frame Supported Bottom Corer," *Ophelia* **10**, 119.

Mc75 McLendon H. R., Stewart D. M., Boni A. L., Corey J. C., McLeod K. W. and Pinder J. E., 1975, "Relationships among Plutonium Contents of Soil, Vegetation and Animals collected on an adjacent to an Integrated Nuclear Complex in the humid Southeastern U.S.A.," *Proc. IAEA Symp. on Transuranium Nuclides in the Environment* p. 347, San Francisco, U.S.A.

No72 Noshkin V. E., 1972, "Ecological Aspects of Plutonium Dissemination in Aquatic Environments," *Health Phys.* **22**, 537.

No75 Noshkin V. E., Wong K. M., Marsh K., Eagle R. and Holladay G., 1975, "Plutonium Radionuclides in the Groundwaters at Enewetak Atoll," *Proc. IAEA Symp. on Transuranium Nuclides in the Environment*, p. 517, San Francisco, U.S.A.

Ta71 Talvitie N. A., 1971, Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange," *Analyt. Chem.* **43**, 1827.

Vi50 Vibe C., 1950, "The Marine Mammals and the Marine Fauna in the Thule District (Northwest Greenland) With Observations on Ice Conditions in 1939–41," *Meddr. Grønland* **150**, 116.

Wa75 Wahlgren M. A. and Marshall J. S., 1975, "The Behaviour of Plutonium and other Long-Lived Radionuclides in Lake Michigan. I. Biological Transport. Seasonal Cycling and Residence Times in the Water Column," *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment*, p. 227, Otaniemi, Finland.

Radiation Botany, 1969, Vol. 9, pp. 357 to 366. Pergamon Press. Printed in Great Britain.

ON THE DIRECT CONTAMINATION OF RYE, BARLEY, WHEAT AND OATS WITH ^{85}Sr, ^{134}Cs, ^{54}Mn AND ^{141}Ce

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Abstract—Radionuclides of Sr, Cs, Mn and Ce were applied as a spray to spring varieties of rye, oats, barley and wheat at four different stages of development.

The Cs and Mn concentrations in the grain were higher than the concentrations of Sr and Ce. The ratios Cs/Sr, Cs/Ce, Mn/Sr, Mn/Ce, and Sr/Ce in the grain increased with the time between spraying and maturity. Field loss of Sr was greater than for the other nuclides applied. The 'half-lives' for the Sr-loss was calculated to about 3 weeks as compared to 4–5 weeks for Mn and Ce and probably even longer for Cs.

Résumé—On a pulvérisé les radionuclides suivants Sr, Cs, Mn et Ce sur les variétés vernales de seigle, d'avoine, d'orge et de froment à quatre stades différents du développement.

Les concentrations de Cs et Mn des grains sont plus élevées que celles de Sr et Ce. Les rapports Cs/Sr, Cs/Ce, Mn/Sr, Mn/Ce et Sr/Ce du grain s'accroissent entre le moment de la pulvérisation et la maturité. La perte de Sr dans le champ est plus élevée que celle des autres nuclides. La 'demi-vie' pour la perte de Sr a été évaluée à environ 3 semaines comparativement à 4–5 semaines pour Mn et Ce et probablement d'avantage pour Cs.

Zusammenfassung—Radionukleotide von Sr, Cs, Mn und Ce wurden in vier verschiedenen Entwicklungsstadien auf Frühjahrsvarietäten von Roggen, Hafer, Gerste und Weizen gesprüht.

In den Körnern waren die Cs- und Mn-Konzentrationen höher als die Konzentrationen an Sr und Ce. Die Verhältnisse der Konzentrationen von Cs/Sr, Cs/Ce, Mn/Sr, Mn/Ce und Sr/Ce in den Körnern erhöhten sich mit zunehmender Dauer zwischen Behandlung und Reife. Die Feldverluste von Sr waren grösser als die der anderen verwendeten Nukleotide. Die Halbwertszeiten für den Sr-Verlust wurden mit etwa 3 Wochen berechnet, für Mn und Ce dagegen mit 4–5 Wochen und für Cs wahrscheinlich mit noch mehr.

INTRODUCTION

THE CONTAMINATION of vegetation with nuclear debris takes place in two ways:[8]

(i) by direct contamination, i.e. adsorption of the debris on the aerial parts of the vegetation often followed by an absorption, and

(ii) by indirect contamination, i.e. absorption through the root system, of radionuclides which have entered the soil.

While all radionuclides in nuclear debris play a role in direct contamination, it is among the long-lived nuclides, mainly ^{90}Sr, and in special cases ^{137}Cs,[2] that absorption from the soil via the roots occurs. During periods with high fallout rates, as in 1962–64, most of the activity (^{90}Sr, as well as other fallout nuclides) in grass and cereals comes from direct contamination. In areas with intensive farming and high calcium content of soil, as in Denmark, the direct contamination of the crops with radiostrontium might be greater than by indirect contamination even when the fallout rate is low. In 1966, when

the fallout rate in Denmark was 2 mCi $^{90}Sr/km^2$ and the accumulated fallout 55 mCi $^{90}Sr/km^2$, nearly half of the ^{90}Sr in Danish grain and milk was due to direct contamination.[1]

As grain products are main contributors of ^{90}Sr and other long-lived radionuclides (e.g. ^{137}Cs, ^{54}Mn and ^{144}Ce) to total human diet in Denmark, the present investigation concentrated on the direct contamination of grain with radionuclides. A small volume of a solution containing ^{85}Sr, ^{134}Cs, ^{54}Mn and ^{141}Ce was applied at various stages of development. The effect of the presence of awns on the contamination, as well as field losses of activity, were also considered.

EXPERIMENTAL METHODS

The experiments were carried out on a clay-loam soil at Risö in the summer of 1967. 132 circular plots with a diameter of 40 cm were seeded with 40 grains on the 5th of May. The species used were R: rye (Petkus), B: barley (Bomi), W: wheat (Koga II), Wa: wheat with awns (No. 2046) and O. oats (Staal). The contaminations were carried out on the following dates.

Experiment I. (30 plots): June 26th, when the ears had emerged in rye but not in the other species.

Experiment II. (30 plots): July 5th, when the ears had emerged in all species except in the wheat variety with awns.

Experiment III. (48 plots): August 1st. Before spraying, the awns were cut from half of all plots with rye, barley, and wheat (with awns), to examine the influence of awns on direct contamination.

Experiment IV. (24 plots): Contaminated just before harvest—barley on August 16th and the other species on September 4th. As in experiment III the awns were cut from half of all plots with rye, barley and wheat.

Three replicates were systematically placed within each experiment. The contamination of a plot was performed as follows: The spraying device (cf. Fig. 1) was placed over the circular plot and the upper cylinder fixed so that the top of the chamber was approx. 20 cm over the top of the foliage. A 10 m^3 activity solution (8 μCi ^{85}Sr, 4μCi ^{134}Cs, 8 μCi ^{54}Mn and 16 μCi ^{141}Ce on August 31st, 1967, with 25 ppm carrier as nitrate of each nuclide) was injected by a syringe into the activity container. Pressurized argon was applied, and the grain received the activity as a fine spray in 5 sec. Afterwards 10 ml distilled water was sprayed to remove residual activity in the syringe and the spraying aggregate. The spraying chamber was removed from the plot and the inner walls of the chamber were washed with ½ l. water, which was collected in a polyethylene bottle for later γ-activity determination. This washing removed nearly 90 per cent of the activity retained on the walls. On the average 9·8 ± 3·5 per cent (1SD) of the 10 ml activity solution applied to each plot was found in the wash-water. After the spraying a wire cage covered with plastic foil was placed over the plot for 2 days to prevent wind and precipitation from removing the activity from the plants before the drops from the spray could dry.

The deposition of activity onto a plane 20 cm below the spray nozzle was tested by spraying filter-paper. This experiment revealed a significantly higher deposition in the center of the plot than in the periphery. To avoid possible consequences of this inhomogeniety all plant material in a plot was included in the measurement sample.

Half of all plots in each experiment, except IV, were harvested 2 days after the contamination to determine the initial retention of radioactivity. In experiment IV the mature samples were identical with the initial samples. Mature as well as initial samples (indicated by indices: 0) were cut just above the soil surface. The samples were dried at 80°C for 24 hr. Initial samples were measured as total plants, except in III, where the grain was measured separately. The mature samples were divided into straw, husks, and grain.* The samples were γ-measured on a lithiumdrifted 2·3 cm^3 planar germanium detector.[4] The samples were measured along with standards of 10 ml spray solution having the same geometry as the samples; the results were reported in percentage of activity of

*Oats and barley had their inner husks, while wheat- and rye-grain had no husks.

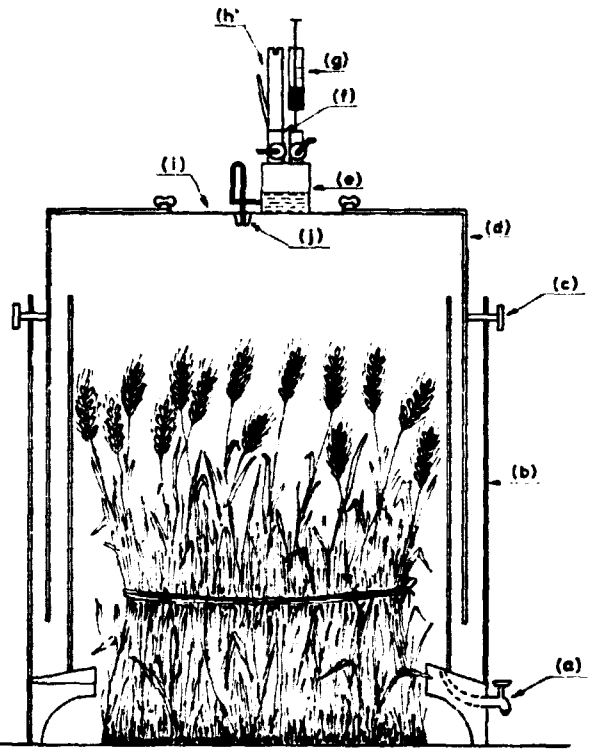

FIG. 1. The spraying chamber. (a) tap for draining-off wash water, (b) the lowermost Plexiglass cylinder, (c) plugs for clamping the cover, (d) the movable Plexiglass cover, (e): stainless steel container for the activity solution, (f) 'Fool proof' branches, (g) injection of activity solution, (h) inlet tube for pressurized air, (e) Plexiglass disk, (j) stainless steel nozzle.

the standard, i.e. the amount sprayed over each plot corrected for decay.

To overcome the difficulties in the statistical analysis which might arise from differences in the growths of the crops, or from variations in the application of the spray, or from local variations in the experiment field, it was decided to study the nuclide ratios. The four nuclides yield six ratios: Cs/Ce, Cs/Sr, Cs/Mn, Mn/Sr, Mn/Ce, Sr/Ce.* The first step in the statistical analysis was a transformation of these ratios to their natural logarithms, as the ratios had a log-normal rather than a normal distribution. Second, a 3-sided analysis of variance was performed on each of the six ratios with the main effects: *Experiments* (I, II, III, IV, and III_0), *species* (R^-, R^+, O, B^-, B^+, W, Wa^-, Wa^+)† and *plant parts* (grain, husks, and straw). The 3-sided analysis of variance was followed by two sets of 2-sided analysis of variance; these included 30 analyses within experiments (main effects: species and plant parts) and 18 analysis within plant parts (main effects: species and experiments).

As the interactions were significant in nearly all analyses of variance the main effects were tested against the interactions only. A difference

*The atomic weights are emitted here and in the following chapter.

†Indices $^-$ and $^+$ refer respectively to the absence or presence of awns before the spraying.

was considered significant if the confidence level was 0·01 or less.

RESULTS AND DISCUSSION

The yields

Tables 1 and 2 show that the yields of the wheat species were lower than the yields of the other species. All yields in the experiment were significantly greater than is usual under field conditions. This might be a result of the great number of plants per unit area in the experiment.

The initial retention

The rather dense growth of the plants resulted in a high initial retention in the crops as shown in Table 3. The spraying of a very small volume of active solution would tend to minimize the dependence of initial retention on species and nuclide. It was, however, possible in the analysis of variance of the initial ratios between pairs of nuclides (cf. Table 5) to demonstrate, that the initial adsorption of Ce in experiments III and IV was less than for the other nuclides.

The retention in mature crops

As the plants varied in size and morphology the activities found in mature crops were expressed as the percentage of the amount initially retained by the crop (cf. Table 4).

Table 1. Dry matter yields of the aerial parts of the plants in grams per plot at harvest. Means of all experiments, ± 1 SE*

Species	Grain	Straw	Husks	Total aerial parts of the plant
Rye	128 ± 5 a	168 ± 6 a	26 ± 1 a	322 ± 13 a
Oats	122 ± 13 a	115 ± 13 b	22 ± 2 a	259 ± 26 b
Barley	118 ± 6 a	126 ± 4 b	24 ± 2 a	268 ± 9 b
Wheat	47 ± 6 b	69 ± 8 c	26 ± 3 a	142 ± 17 c
Wheat with awns	28 ± 3 c	76 ± 6 c	18 ± 2 a	122 ± 10 c

*The experiments did not differ significantly from each other in yields. Results with the same letters in a column are not significantly different ($P < 0{\cdot}01$).

Table 2. Preharvest dry matter yields of the aerial parts of the plants in grams per plot

	Exp. I_0	Exp. II_0	Exp. III_0	
	Total yield	Total yield	Grain	Total yield
Rye	173	150	71	301
Oats	71	75	99	219
Barley	131	140	120	295
Wheat	40	49	20	111
Wheat with awns	53	52	12	122

MIDDLETON[5] found that wheat sprayed before ear emergence contained 0·04–0·12 per cent of the initially retained ^{90}Sr in the grain and 5·2–10·3 per cent of the initially retained ^{137}Cs. In samples sprayed after ear emergence, he found 0·86–1·79 per cent ^{90}Sr and 3·0–10·2 per cent ^{137}Cs. Table 4 shows that these results are in reasonable agreement with the corresponding percentages in the present experiment.

Analysis of variance on the nuclide ratios showed that in most cases there were no significant differences between species as regards the

Table 3. Activity initially retained on the total crops expressed as the percentage in the spraying solution

Species	Nuclide	Exp. I	Exp. II	Exp. III	Exp. IV
Rye with awns cut off	Ce	—	—	44·4	51·9
	Sr	—	—	55·1	52·9
	Mn	—	—	57·0	56·0
	Cs	—	—	63·7	55·8
Rye (with awns)	Ce	82·5	61·3	40·8	48·7
	Sr	81·0	57·3	48·0	55·7
	Mn	77·5	56·6	47·8	57·0
	Cs	79·1	58·7	51·4	55·7
Oats	Ce	66·1	61·5	41·8	41·7
	Sr	61·2	62·9	46·1	53·3
	Mn	59·5	61·9	44·1	48·8
	Cs	59·5	64·8	48·1	49·7
Barley with awns cut off	Ce	—	—	49·4	60·8
	Sr	—	—	56·8	74·8
	Mn	—	—	58·8	77·6
	Cs	—	—	58·8	77·0
Barley (with awns)	Ce	95·7	79·2	49·5	65·5
	Sr	91·6	79·5	58·3	70·8
	Mn	97·6	82·2	58·4	72·0
	Cs	87·5	79·3	62·9	73·4
Wheat (without awns)	Ce	50·8	51·0	30·1	43·1
	Sr	47·0	49·8	34·9	49·5
	Mn	48·8	44·8	35·2	49·0
	Cs	49·4	51·9	39·8	49·7
Wheat with awns cut off	Ce	—	—	43·1	47·4
	Sr	—	—	49·3	55·0
	Mn	—	—	47·8	45·4
	Cs	—	—	52·8	53·8
Wheat (with awns)	Ce	80·7	56·4	41·5	50·6
	Sr	79·3	63·9	50·8	57·5
	Mn	78·8	57·5	50·9	56·6
	Cs	76·1	55·4	53·6	56·8

six nuclide ratios. Consequently, the means of all species were reported (cf. Table 5).

Grain. All ratios except Cs/Mn, showed significantly higher values in exp. I and II than in III and IV, and all ratios in I and II were significantly higher than the initial ratios. This confirms the expected order: Cs > Mn > Sr > Ce for absorption and translocation of these elements.

The longer the times between contamination and harvest, the greater were the ratios.

Husks. The ratios in husks were significantly less than in grain, and in accordance with the translocation pattern for the 4 nuclides. Table 5 furthermore, shows that Cs/Ce, Cs/Sr, Cs/Mn, and Mn/Sr in husks were greater in exp. I and II than in III and IV, and that the ratios in I and II were significantly higher than the initial ratios in the total plant. This indicates that the translocation of Cs from straw to husk occurs preferentially as compared to the other nuclides.

Straw. The ratios in straw were, as expected,

Table 4. Percentage of initially retained activity in total crops in grain, husks and straw

Species	Nuclide	Experiment I			Experiment II			Experiment III			Experiment IV		
		Grain	Husks	Straw	Grain	Husks	Straw	Grain	Husks	Straw	Grain	Husks	Straw
Rye with awns cut off	Ce	—	—	—	—	—	—	1·4	35·6	20·1	4·1	27·0	68·9
	Sr	—	—	—	—	—	—	6·3	11·4	13·5	4·3	31·4	64·3
	Mn	—	—	—	—	—	—	11·9	12·8	16·5	3·8	31·1	65·1
	Cs	—	—	—	—	—	—	19·4	8·1	11·7	6·5	30·8	62·7
Rye (with awns)	Ce	0·1	3·0	18·4	0·3	10·7	19·6	1·6	31·9	21·5	3·5	44·3	52·2
	Sr	0·5	1·1	7·8	1·6	3·0	9·2	6·7	11·4	17·6	4·9	41·6	53·5
	Mn	7·6	2·1	13·6	10·7	4·4	10·3	12·7	13·3	22·1	4·2	41·9	53·9
	Cs	14·6	2·3	12·1	24·3	4·8	12·0	27·1	10·1	15·5	7·7	40·6	51·7
Oats	Ce	0·1	0·5	29·3	0·1	1·9	16·8	1·6	20·0	24·6	4·2	50·0	45·8
	Sr	0·2	0·3	9·3	0·4	0·7	6·3	4·4	12·3	13·2	4·0	43·8	52·2
	Mn	7·0	2·1	23·3	4·7	1·9	12·4	9·5	13·8	21·2	4·7	48·0	47·3
	Cs	11·0	2·8	16·8	10·0	2·0	8·3	16·4	6·6	14·4	6·5	46·6	46·9
Barley with awns cut off	Ce	—	—	—	—	—	—	11·9	26·0	45·5	8·2	27·0	64·8
	Sr	—	—	—	—	—	—	13·9	17·4	53·5	7·1	25·4	67·5
	Mn	—	—	—	—	—	—	12·6	20·5	56·0	6·6	24·1	69·3
	Cs	—	—	—	—	—	—	15·5	10·5	38·5	9·5	23·5	67·0
Barley (with awns)	Ce	0·2	3·2	18·5	0·8	10·4	6·5	7·8	30·8	25·1	6·0	46·9	47·1
	Sr	0·2	1·0	10·1	0·7	3·3	9·5	8·5	20·8	17·5	5·1	41·1	53·8
	Mn	4·4	2·8	24·7	8·4	7·5	21·2	8·6	26·1	25·7	4·9	40·3	54·8
	Cs	9·0	3·1	14·4	15·5	4·3	16·3	13·5	10·2	14·3	7·4	39·8	52·8
Wheat (without awns)	Ce	0·04	0·8	29·8	0·06	5·0	17·7	0·2	28·0	43·2	0·5	52·2	47·3
	Sr	0·2	0·9	15·9	0·8	4·2	8·2	2·3	23·6	24·1	0·3	54·2	45·5
	Mn	3·2	1·8	22·5	3·9	5·1	13·8	4·0	21·1	34·7	0·3	52·6	47·1
	Cs	11·9	6·6	16·9	10·8	5·6	7·7	8·4	13·0	17·5	1·1	55·0	43·9
Wheat with awns cut off	Ce	—	—	—	—	—	—	0·6	14·0	56·9	0·3	27·8	71·9
	Sr	—	—	—	—	—	—	1·0	12·2	24·8	0·3	29·7	70·0
	Mn	—	—	—	—	—	—	2·4	11·3	35·4	0·3	36·3	63·4
	Cs	—	—	—	—	—	—	2·9	7·2	16·8	1·0	30·9	68·1
Wheat (with awns)	Ce	0·02	0·2	25·6	0·02	0·4	26·5	0·07	18·4	34·4	0·3	41·8	57·9
	Sr	0·08	0·2	7·1	0·09	0·5	8·8	0·9	11·5	18·6	0·4	43·1	56·5
	Mn	1·7	0·8	15·0	2·3	0·9	16·8	2·4	11·7	26·9	0·2	41·8	58·0
	Cs	4·0	2·8	12·8	3·9	2·7	13·0	3·6	11·8	15·0	1·1	43·2	55·7

Table 5. Mean ratios for all species with a summary of the results of the analysis of variance

Ratio	Experiment	Grain	Husks	Straw	Total plant initially
Cs/Ce	I	a185α	a6·3β	b0·58γ	b0·94
	II	a130α	b1·9β	b0·55β	b1·00
	III	b30α	c0·52β	b0·73β	a1·27
	IV	c2·5α	c1·14β	a1·16β	ab1·15
	III_0	c2·9α		a1·21β	
Cs/Sr	I	a65α	a7·5β	a1·46γ	b0·99
	II	b26α	b2·6β	a1·33β	b1·00
	III	c3·3α	c0·75β	b0·94β	a1·09
	IV	c2·3α	c1·00β	b0·99β	b1·01
	III_0	c3·4α		b1·01β	
Cs/Mn	I	a2·3α	a2·1α	bc0·74β	a0·98
	II	a2·3α	ab1·42α	bc0·82α	a1·03
	III	a1·8α	c0·68β	c0·66β	a1·08
	IV	a2·7α	bc1·00β	ab1.02β	a1·03
	III_0	a2·9α		a1·20β	
Mn/Ce	I	a75α	a2·5β	b0·84γ	b0·96
	II	a61α	a0·97β	b0·70β	b0·98
	III	b17α	a0·79β	a1·08β	a1·17
	IV	c1·01α	a1·14α	a1·17α	a1·13
	III_0	c1·27α		a1·18β	
Mn/Sr	I	a29α	ab3·8β	a2·1β	a1·01
	II	a13α	bc1·9β	ab1·7β	a0·97
	III	b1·9α	cd1·13β	bc1·44β	a1·00
	IV	c0·89α	d1·00α	c1·00α	a0·98
	III_0	bc1·5α		c0·99β	
Sr/Ce	I	a3·3α	a0·67β	b0·41β	b0·96
	II	a6·0α	a0·57β	b0·43β	b1·01
	III	a8·1α	a0·72β	ab0·76β	a1·17
	IV	b1·15α	a1·15α	a1·17α	a1·15
	III_0	b0·84β		a1·19α	

Ratios with the same roman letter in a column are not significantly different ($P < 0.01$). Ratios with the same greek letter in a row are not significantly different.

usually less than in husks. In exp. III and IV there were, however, no differences between the two plant parts. The straw ratios in these experiments were close to the initial ratios in total plants. In exp. I and II the ratios in straw were less than the initial ratios in total plants, except for Cs/Sr and Mn/Sr. This is explained by the differences in the transport of the nuclides to the grain (and husks), as well as by the differences in the nature of field loss (cf. Fig. 3).

Total plant. Figure 2 shows (as an example) the relative distribution of the four nuclides within the rye plant in the different experiments. Each nuclide was given a sector of 90° irrespective of its actual contribution to the total activity of the plant. It appears that the Cs and Mn found in the grain increase according to the length of the period between contamination and harvest; this, at the expense of their levels in straw and husks. It also seems that Sr and

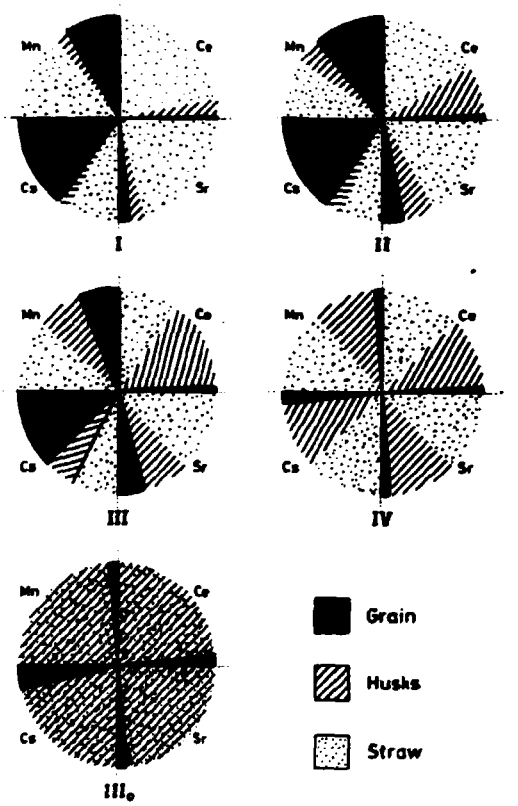

FIG. 2. The relative distribution of activity between grain, straw and husks in rye (R^+).

Ce mostly stick to the straw in exp. I, indicating little or no translocation of these nuclides to the grain. The figure also shows that husks, especially in exp. III and IV, contain a considerable part of the total activity, although the husks amount only to less than one-tenth of the total weight of the aerial parts of the plant (cf. Table 1). This preferential uptake in the husks might be a result of the method of spraying.

Difference between species. The analysis of variance showed that the Cs/Ce and Cs/Sr ratios in grain and the Cs/Ce ratios in husks were higher in wheat than in barley, especially in exp. I and II (cf. Table 4). The reason might be that the ears of wheat in I and II were less developed than the ears of barley.

The Mn/Ce ratio in straw was significantly higher in barley than in the other species (cf. Table 4). This might be a result of the shorter time available for the translocation of Mn from straw to grain in the case of barley, as this species was harvested at an early date.

In exp. I the Mn/Sr ratio was significantly higher in oats than in rye. This is explained by the fact that rye was the only species in exp. I where the ears had appeared, and thus the only species which absorbed Sr in appreciable quantities in the grain and husks.

Table 4 indicates that grain from rye with awns showed higher activity levels than grain from rye from which awns had been cut. The opposite was the case for barley. The reason

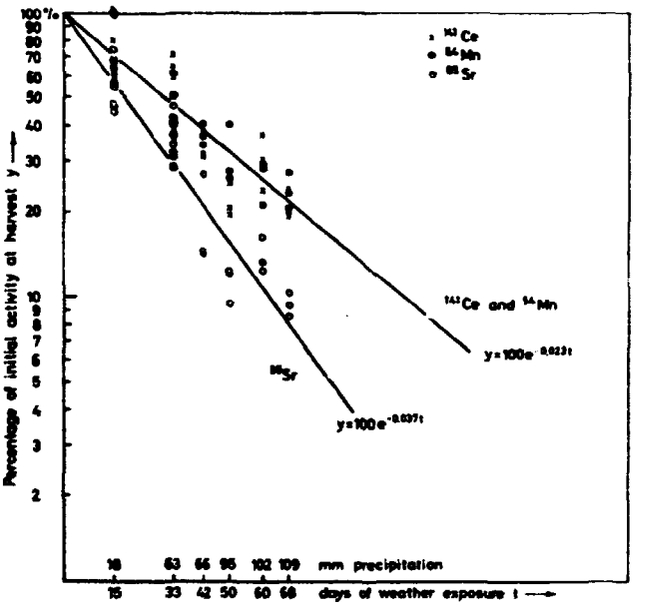

FIG. 3. The field loss of ^{85}Sr, ^{141}Ce and ^{54}Mn in the aerial parts of rye (R/R_0) and barley (B/B_0).

for this different behaviour of rye and barley might be that the removal of the awns from barley leaves an open wound in the innerhusks, thus rendering the barley grain more susceptable to contamination. As rye has no innerhusks this effect does not play any role. It seems that, in this case, the presence of awns is the dominating factor.

Interactions. In all 3-sided analyses of variance on the nuclide ratios the two-factor interaction between experiment and plant part was significant. The 2-sided analysis of variance showed, in most cases, significant interactions between experiment and species, and between species and plant part. The interactions are explained by the fact, that the different species differed in development especially in experiment I and II (ears emerged or still hidden) and thus reflected different abilities to absorp the different nuclides.

Loss of activity from total plants

The Sr levels in grain as well as in straw and husks were less than the levels of Mn, although the initial uptake of these nuclides in the total plants, was nearly the same (cf. Tables 4 and 5). The field loss was thus greater for Sr than for Mn. MIDDLETON[6] was not able to prove any significant difference between the removal times of various nuclides in cabbage (for ^{54}Mn, ^{89}Sr, ^{95}Zr, ^{106}Ru, ^{131}I, ^{137}Cs, ^{144}Ce) nor in grass (for ^{89}Sr, ^{137}Cs, ^{144}Ce).

Figure 3 shows the mean ratios between harvest activity and initial activity in the aerial parts of rye and barley. (Oats and wheat were excluded in this comparison due to significantly greater variances than for rye and barley). The figure shows that Ce and Mn with a half-life of 4–5 weeks were lost from the plants, while the half removal time for Sr was approx. only 3 weeks. The behaviour of Cs (not indicated in the figure) seems to be different from the other nuclides. There might be a double exponential field loss of Cs from rye and barley: one rather fast and one more slowly. MIDDLETON[6] found that ^{137}Cs was retained to a somewhat greater extent than the other nuclides in his cabbage experiment, but he

could not prove any statistically significant differences.

In the present experiment it was of statistical significance that Sr was more easily removed from the aerial parts of rye and barley than Ce and Mn. Middleton found in his grass experiment that the removal of ^{89}Sr, ^{137}Cs and ^{144}Ce could be described by a simple exponential curve of the form $y=e^{-kx}$, where x is the number of days after spraying and y the percentage of the initial deposition which remained on the herbage. The value of the constant k he found equal to 0·05 with a standard error of $\pm 0{\cdot}02$. In a later experiment, MILBOURN[7] found for ^{89}Sr in grass $k=0{\cdot}054 \pm 0{\cdot}005$. The values for k in the present experiment were 0·037 for Sr (SE: 0·002) and 0·023 for Ce and Mn (SE: 0·002).

The faster removal of Sr than of Ce and Mn from the plants might be explained by a greater solubility of the *absorbed* Sr than of the *absorbed* Ce and Mn. To verify this, 4 grain samples, one of each species, from experiment III, were washed (four months after the harvest) with distilled water (4 vol. H_2O/1 vol. grain) for 90 sec during mechanical stirring. Of the Ce, Cs and Mn, 1·5 per cent were removed and of the Sr 6·5 per cent, by this washing. This supports the findings in the present field experiment.

Indirect contamination of the crops through the root system has not been considered in this experiment, but is assumed not to be of any significance.

SUMMARY AND CONCLUSIONS

The direct contamination of cereal crops with Cs and Mn follow similar pictures: both nuclides (Cs somewhat more readily than Mn) are translocated within the plant and the activity levels in the grain relative to that in the vegetative parts of the plants increase with time from contamination to harvest. Ce, and to some extent Sr, shows the opposite picture: after the contamination the grain content of Ce will decrease relative to the straw. Sr and Ce show maximum grain levels, if the contamination has occurred during the last month before harvest. The concentration of Ce and Sr in grain are relatively low as compared to Mn and Cs. Sr is, unlike Ce, transported, although in minor amounts, from other aerial parts of the plant to the grain. Hence the grain contained more Sr than Ce, while the opposite was the case for the straw.

The field loss of Sr in the aerial parts of the plants was greater than the loss of the other nuclides.

The experiment did not give an unambiguous answer to the question of the importance of awns in direct contamination, and it did not give sufficient information as to why rye grain in survey studies[2] shows higher activity than other species.

Acknowledgements—The author gratefully acknowledges the assistance of A. ANDERSEN and V. HAAHR from the Agricultural Research Department at Risø, who helped in the planning of the experiment and the discussion of the results: of B. LARSEN, A. HOLM PEDERSEN and all the others in the Health Physics Department, who collected, processed and analysed the plant samples.

REFERENCES

1. AARKROG A. and LIPPERT J. (1967) Environmental Radioactivity in Denmark 1966. Danish Atomic Energy Commission, *Risø Report* 154.
2. AARKROG A. (1968) On the variation of the levels of ^{90}Sr and other fallout nuclides in the grain of rye, barley, wheat and oats. pp. 1047–1063. *Proc. Intern. Congr. Intern. Radiation Protection Assoc., 1st*, Pergamon Press, Oxford.
3. FREDERIKSSON L. (1963) Studies on plant absorption of Sr-90 and Cs-137 from some subtropical and tropical soils. *Försvarets Forskningsanstalt, FOA 4 Rapport*, A 4319–4623.
4. LIPPERT J. (1968) Some applications for semiconductor detectors in Health Physics. pp. 263–269. *Proc. Intern. Congr. Intern. Radiation Protection Assoc. 1st*, Pergamon Press, Oxford.
5. MIDDLETON L. J. (1959) Radioactive strontium and caesium in the edible parts of crop plants after foliar contamination. *Intern. J. Radiation Biol.* 1, 387–402.
6. MIDDLETON L. J. and SQUIRE H. M. (1961) Comparison of the retention of various nuclides by cabbage. *Agr. Res. Council Radiobiol. Lab. Rept. ARCLR* 5, 50.
7. MILBOURN G. M. and TAYLOR R. (1965) The contamination of grassland with radioactive Strontium-I initial retention and loss. *Radiation Botany* 5, 337–347.
8. RUSSELL R. S. (1966) *Radioactivity and Human Diet*, pp. 89–104, Pergamon Press, Oxford. 532 pp.

Radiation Botany, 1971, Vol. 11, pp. 463 to 472. Pergamon Press. Printed in Great Britain.

DIRECT CONTAMINATION OF BARLEY WITH ^{51}Cr, ^{59}Fe, ^{58}Co, ^{65}Zn, ^{203}Hg AND ^{210}Pb

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AARKROG A. and LIPPERT J. *Direct contamination of barley with ^{51}Cr, ^{59}Fe, ^{58}Co, ^{65}Zn, ^{203}Hg and ^{210}Pb.* RADIATION BOTANY **11**, 463–472, 1971.—A study of barley sprayed at six different stages of development with radionuclides of Cr, Fe, Co, Zn, Hg and Pb. The initial retention followed the equation: $IR = 1 - e^{-0.31\,g.cm^{-1}}$, where g is the dry weight of the herbage in a plot in grammes, and cm the height of the plants at spraying.

The field loss depended on the growing rate and the development of the plants and thus varied throughout the growing period. The field loss coefficient λ was 0·054 days^{-1} in the first part of the period and 0·017 days^{-1} in the second part. If the whole growing season is considered, the loss of activity in per cent is about equal to the time in days from the spraying to harvest.

Zn and Co showed the highest concentrations in the grain. The translocation of Cr, Pb and Hg within the plant was small. Fe was translocated to the grain as was Zn, but to a far less extent.

INTRODUCTION

IN AN earlier paper[1] the direct contamination of rye, barley, wheat and oats with ^{85}Sr, ^{134}Cs, ^{54}Mn and ^{141}Ce has been described. The four nuclides were chosen as representative of the most common long-lived radionuclides in fall-out from nuclear weapons testing. Cs and Mn showed an evident translocation, whereas Sr and especially Ce only moved little within the plants.

The aim of the present experiment was to investigate the behaviour of six other nuclides. Cr, Fe, Co and Zn were chosen because they are common corrosion products in nuclear power plants[5] and as regards the three last named occasionally also occur in fall-out.[14] Hg and Pb were selected because they belong to the important nonradioactive pollution products, and besides ^{210}Pb is a constituent of natural fall-out.[8]

Information on intra plant transport of corrosion products and heavy metals could be useful in connection with assessments of food-chain movements and radioecological hazards.

Only barley was considered in the present experiment because it has previously[1] been shown that an experimental comparison between the different species was not possible with the experimental technique we used.

EXPERIMENTAL METHODS

The experiment was carried out in the same area and after the same principles as used in 1967.[1] The experimental field was divided into forty-eight circular plots made up by eight rows and six columns. Each row was an experiment and was designated: I, II–VI, A, O, to minimize cross-contamination between experiments, they were arranged systematically according to increasing degree of contamination (I was lowest and VI highest at harvest). Table 1 shows the data for the experiments.

The spraying chamber used in 1967[1] was modified; the upper part was unaltered, but we replaced the walls by a disposable polyethylene bag which was renewed after each

spraying; 5–10 per cent of the dosed activity were found on the bags. The bags were one metre long with a diameter of approx. forty-eight cm and had no bottom. They were stretched by three rings and carried by a rack. Apart from this the spraying procedure was the same as in 1967.

After each spraying (cf. Table 1), which was performed in the order: experiment O (one plot), experiment A (six plots) and experiment Roman numeral (six plots), the area was protected against rain for 24 hr. After this drying period we harvested every other plot in the Roman numeral experiments to determine the initial retention.

The plant material was dried at 80°C for 24 hr and divided into grain, husks and straw in a glove box. We measured the radioactivity on a 30 cm³ Ge(Li) detector connected to a 1024-channel Hewlett Packard analyser and a H.P. 9100 B Calculator, which permitted a rapid and accurate calculation of the six nuclides in the samples. The following photo peaks were used: ^{210}Pb: 47 keV*, ^{57}Co: 122 keV, ^{203}Hg: 279 keV, ^{51}Cr: 320 keV, ^{65}Zn: 1110 keV and ^{59}Fe: 1290 keV.

RESULTS AND DISCUSSION

The initial retention: IR

IR is the ratio between the activity found in the herbage from the plot and the amount of activity dosed. IR was expected to be related to the surface of the plants at spraying time, and in fact we found from table 2 that

$$IR = 1 - e^{-0.31\, g\,cm^{-1}} \qquad (r = 0.9628),$$

where g is the dry-matter weight, cm the height of the plants as shown in Table 1, and r the coefficient of correlation.

Contrary to the 1969 experiment[1] the initial retention in the present experiment was on the average only 80 per cent calculated for the same stage of development. This could be a result of differences in the drying conditions.

Table 2 shows that the uptake of Hg was approx. two-thirds of that of the other nuclides, probably on account of an easier evaporation of this nuclide. The other nuclides did not differ significantly in initial retention.

Table 1. Experimental data

Experiment	Spraying date	Height of plants, cm	Dry matter weight of herbage: grammes ± 1 SE at spraying	Spraying solution	Remarks
I	May 28	13	3·4 ± 0·8	U	
II	June 15	33	37 ± 8	U	
III	June 25	50	69 ± 3	U	Start of ear emergence
IV	July 9	79	182 ± 33	U	The ears fully emerged
V	July 27	79	277 ± 6	U	
VI	Sept. 2	79	317 ± 6	U	
A	May 28, June 15, 25 July 9, 27, Sept. 2	—	—	U/6	All plots were sprayed each time
O	May 28, June 15, 25 July 9, 27, Sept. 2	—	—	H_2O	Only one plot was sprayed each time

The spraying solution U contained on April 1, 1970: 5 mCi ^{51}Cr, 1·25 mCi ^{59}Fe, 2·5 μCi ^{57}Co, 50 μCi ^{65}Zn, 125 μCi ^{203}Hg, and 25 μCi ^{210}Pb per 10 ml solution with 25 ppm carrier as nitrate of each nuclide. Solution U/6 consisted of one volume U and five volumes H_2O, 10 ml being sprayed on each date.

The grain was seeded on April 30 and harvested on Sept. 3, 1970.

In the mature plants 58 per cent were grain, 11 per cent husks and 31 per cent straw on dry-weight basis.

*The counting efficiency for the 47 keV photo peak from ^{210}Pb was so low that a possible ^{210}Pb content in the weakest samples (e.g. grain from experiments I, II and III) was not measureable.

Table 2. Activity retained in crop immediately after spraying, expressed as per cent of total activity in the initial spraying solution

Nuclide	Expt. I	Expt. II	Expt. III	Expt. IV	Expt. V	Expt. VI	Mean
Pb	8·2±0·5	26± 2	48±7	61±7	63±2	71±6	46
Co	3·6±0·2	29± 2	51±7	66±6	70±2	76±1	49
Hg	2·4±0·1	37± 4	21±4	41±4	48±2	62±2	35
Or	3·7±0·3	36± 4	48±6	61±5	67±3	71±2	48
Zn	3·8±0·3	60±10	45±6	59±5	62±2	68±1	50
Fe	3·6±0·3	49± 8	47±6	60±4	65±2	67±2	49
Mean	4	40	43	58	63	69	

The error term is the SE of the mean of triplicates.

The content in mature crops

Experiment A, which represented repeated contamination throughout the growing season, showed more or less the same distribution of Pb, Hg, Cr and Fe: within the plant just about 10 per cent in grain, 30–35 per cent in husks and the remainder in straw. Co and Zn showed 30–40 per cent in grain, 25–30 per cent in husks and the remainder in straw. As compared with the weights of the three parts of the plant (cf. Table 1) it is evident that the grain receives less and the husks more than was to be expected from the weight alone. This is, however, only the case when contamination is performed as in experiment A (cf. Fig. 1).

When we compared the levels in experiment A with mean of the six Roman numeral experiments: (Σ/6), the mean ratio between A and Σ/6 of all nuclides and parts of plants was 0·94±0·05 (1 SE), i.e. not significantly different from 1. The main reason for the mean coming out as less than 1 was Hg, which for all plant parts showed lower levels for the plots sprayed with the diluted solution. In our opinion this can be ascribed to the dilution of the activity without maintenance of the carrier concentration, which will displace the equilibrium

$$Hg + Hg^{++} \rightleftharpoons Hg_2^{++}$$

to the left and thus increase the possibility of evaporation of Hg. The lower Hg levels in experiment A could, however, also be accounted for by the repeated sprayings in that experiment which might have rubbed off some of the deposited Hg.

In this comparison two factors were confounded: the concentration of the spraying solutions and the additivity of successive sprayings. Thus we cannot (except for Hg) preclude an effect from dilution of the spraying solution, but it seems highly improbable to us that the two factors should exactly neutralize each other.

Indirect contamination

In experiment I the barley was allowed to regenerate on the three plots used for initial samples. The regenerated plants were harvested along with the other samples, and the mean levels in this experiment I′ are shown in Table 3.

The total plants from experiment I′ (corrected to the same yield as I) showed 80 per cent Co, 50 per cent Hg, 20 per cent Zn and 20 per cent Fe of the respective nuclide levels in mature total plants in experiment I. In the case of grain the percentages of the mature grain levels were: Co: 1·5, (Hg: 65)*, Zn: 20, and Fe: 25. Root uptake (and stem base absorption) could thus not be completely neglected in experiment I, but was usually less than a few per cent of the activity level in the mature plants in the other experiments (note that the figures in Table 3 are the percentages of initially retained activity in total crops, i.e. in the case of experiment I′ the figures are the initial levels from experiment I in Table 2).

Field loss. In the 1967 experiment[1] we found

*On account of the very low Hg levels in grain the percentage is very approximate.

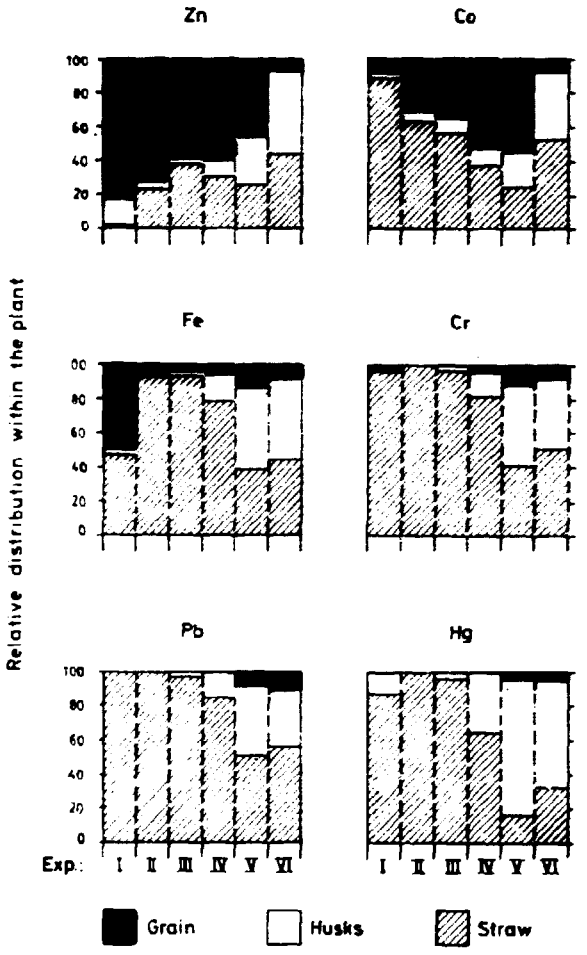

FIG. 1. The relative distribution of the activity within the plants for each of the six experiments.

that Sr was removed from the plants with a half-life of about three weeks, i.e. the coefficient of field loss was $\lambda = 0{\cdot}037$ days^{-1}. Ce and Mn were lost with a half-life of four to five weeks ($\lambda = 0{\cdot}023$ days^{-1}) and Cs probably a little more slowly. The present experiment lasted for a longer period (97 days against 68 in 1967), and showed a double exponential field loss.

The field loss was greatest during the first part of the growing season. Later, when the growth rate became slower, the field loss decreased cf. Table 4. This observation is in agreement with that of CHADWICK *et al.*,[9] who found the field loss to be greater in the summer (half-life ~ 19 days) than in winter (half-life 49 days) when the growth of herbage is slow.

A further examination of the field loss for barley reveals a relationship that is closer to a linear than to an exponential expression. The mean loss of activity expressed as the percentages of the activity initially deposited on the plants (i.e. 100 minus the figures in the last column in Table 4) was nearly equal to the number of days from spraying to harvest (cf. Fig. 2).

Table 3. Per cent of initial activity retained in mature grain, husks and straw

		Expt. I	Expt. II	Expt. III	Expt. IV	Expt. V	Expt. VI	Expt. A	Expt. I'
Grain	Pb	—	—	—	0·2	5 ±1	8·2 ±0·8	4·2	—
	Co	0·43 ±0·04	3·4 ±0·3	4·6 ±0·7	18 ±3	29 ±2	6·6 ±0·1	16	0·006 ±0·002
	Hg	0·06 ±0·03	0·007 ±0·002	0·11 ±0·04	0·6 ±0·1	4·2 ±0·4	8·0 ±0·3	2·0	0·04 ±0·02
	Cr	0·02 ±0·01	0·05 ±0·01	0·34 ±0·05	1·5 ±0·2	6·5 ±0·8	7·4 ±0·3	3·6	—
	Zn	11 ±2	8 ±1	15 ±2	29 ±4	32 ±2	7·6 ±0·1	19	2·2 ±0·8
	Fe	1·2 ±0·02	0·9 ±0·2	1·5 ±0·2	2·0 ±0·3	7·5 ±0·5	9·2 ±0·4	4·2	0·3 ±0·1
Husks	Pb	—	—	0·54 ±0·09	5 ±1	22 ±2	25 ±3	15	0·7 ±0·4
	Co	0·1 ±0·1	0·57 ±0·08	1·0 ±0·2	3·4 ±0·4	10·7 ±0·7	33 ±1	14	0·03 ±0·03
	Hg	0·6 ±0·4	0·06 ±0·02	1·3 ±0·4	11 ±2	58 ±4	78 ±5	19	—
	Cr	—	0·04 ±0·01	1·1 ±0·2	6 ±1	25 ±2	38 ±3	15	—
	Zn	0·7 ±0·2	0·42 ±0·08	0·9 ±0·2	3·8 ±0·7	21 ±2	49 ±3	14	0·12 ±0·03
	Fe	0·05 ±0·01	0·07 ±0·01	1·0 ±0·1	5 ±1	26 ±2	17 ±3	14	—
Straw	Pb	—	13 ±2	16 ±2	26 ±9	28 ±8	44 ±5	25	—
	Co	4 ±2	6·5 ±0·9	7 ±1	12 ±3	12 ±3	42 ±2	20	3·7 ±0·8
	Hg	4 ±1	12 ±1	30 ±6	21 ±7	22 ±6	41 ±3	13	2·3 ±0·6
	Cr	0·6 ±1	27 ±3	39 ±5	30 ±7	21 ±5	42 ±3	24	—
	Zn	1·6 ±0·5	2·8 ±0·5	10 ±2	15 ±4	17 ±4	43 ±3	18	0·2 ±0·1
	Fe	1·1 ±0·2	15 ±3	32 ±5	25 ±6	21 ±4	44 ±3	22	0·13 ±0·05

The error term is the SE of the mean of triplicates. All samples were corrected for the background activity found in expt. O. The initially retained activities in expt. A were estimated as the corresponding mean levels in Table 2. The percentage for expt. I' were based on the initial retention in expt. I, and the percentage were calculated for "normal" yield, i.e. a yield approx. 50 per cent higher than found in expt. I'.

Table 4. Percentage of initial activity remaining in total plant at harvest

Expt.	Days from harvest	mm precipitation until harvest	Percentage of initial activity at harvest in total plants (±1 SE)						Mean ± 1 SE
			Pb	Co	Hg	Cr	Zn	Fe	
I	97	87	~0	4·7±1·5 (1·0)	5·1±1·5 (2·8)	0·6±1·5	13·5±2 (11·1)	2·3±0·3 (1·9)	4·4±2 (2·6)
II	79	86·5	13·3±1·5	10·4±0·9	12·0±1·4	27 ±3	11·6±2	16·5±2·7	15 ±3
III	69	80	16·9±2·6	12·5±1·8	31·6±5·8	40 ±5·3	25·6±7	34·8±5	27 ±4
IV	55	57	30·9±9·2	33·8±5·1	32·6±7·2	36·8±7·5	48·2±6·6	32·2±6·1	36 ±3
V	37	34	55 ±8	52 ±4	84 ±8	53 ±6	69 ±5	54 ±6	61 ±5
VI§	0	0	78 ±8	82 ±2	127 ±7	87 ±5	100 ±5	91 ±5	94 ±7
I–V		λ days^{-1}	0·035‡	0·042	0·046	—	0·032	0·048	0·0425 (0·050)
		t1/2 days	20	17	15	—	22	14	16 (14)
		r	0·9965	0·9869	0·9735	—	0·9309	0·8902	0·9660 (0·940)
IV–VI		λ days^{-1}	0·016	0·016	0·022*	0·014†	0·013	0·015*	0·0166 0·0182*
		t1/2 days	44	48	32	51	54	45	42 38
		r	0·9436	0·9837	0·9420	0·9689	0·9825	0·9636	0·9679 0·9788

The figures in brackets (in expt. I) are the field loss corrected for indirect contamination as found from expt. I′ (cf. Table 3).
*Calculated from expts. III–VI.
†Calculated from expts. II–VI.
‡Calculated from expts. II–V.
§In expt. VI the theoretical percentage is 100 i.e. the ratio between the activity in (grain + husk + straw) from three of the six plots and in total plants from the other three plots, however, owing to sampling and measuring errors this did not hold true.

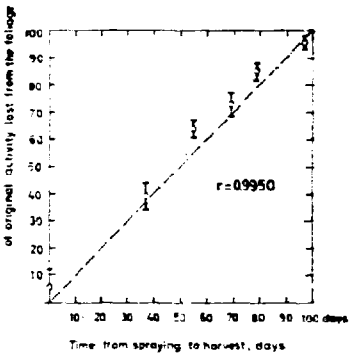

FIG. 2. The mean field loss (±1 SE) of Pb, Co, Hg, Cr, Zn and Fe from barley during the growing season (loss in per cent = days from spraying to harvest).

There was little rain during the first part of the experiment (cf. Table 4), but nevertheless the field loss was greatest in this period. MOORBY *et al.*[11] have suggested that the considerable loss of activity found in rapidly growing plants, even under dry conditions, might be a result of the shedding of wax from the leaves.

The half-lives measured in the 1967 experiment corresponded to the half-lives measured in the second period of the present experiment, which was to be expected as the 1967 experiment was carried out only during the last two-thirds of the growing period, i.e. the 1967 experiment corresponded to experiments III–VI of the present experiment.

Distribution within the plants

Grain. The order of activity levels in grain (cf. Fig. 3) was: Zn > Co > Fe > Cr > Pb ~ Hg. From a regression analysis we found that the Co/Zn ratio in grain from experiments I–V followed the equation

$$(Co/Zn_{grain}) = 0{\cdot}32 g{\cdot}cm^{-1} - 0{\cdot}095 \quad (r = 0{\cdot}9932) \qquad (2)$$

where g and cm were taken from Table 1 [cf. also equation (1)], and the activity ratio was calculated from Tables 2 and 3. Since initial retention was approximately the same for Zn and Co, equation (2) illustrates that Zn is translocated more easily than Co from other parts of the plant to the grain (cf. Fig. 1). The

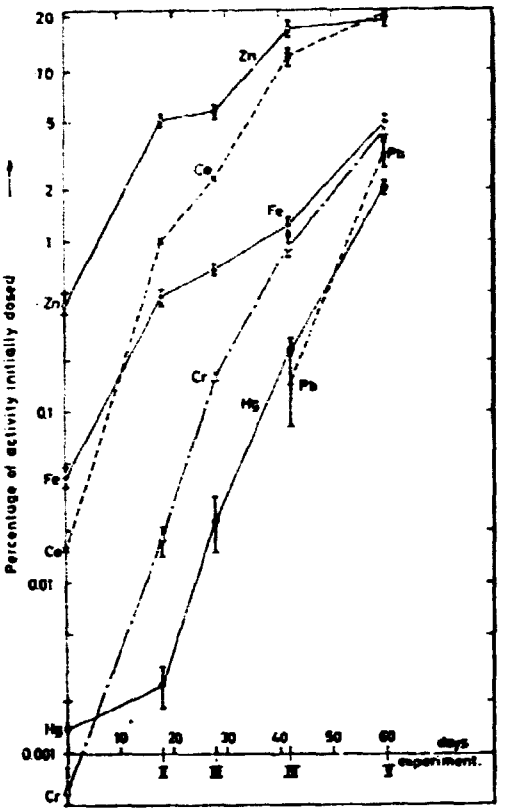

FIG. 3. The levels in mature grain (expressed as percentages of the activity in the 10 ml spraying solution) as a function of spraying time (±1 SE indicated). Pb was not measureable if the level was below 0·05 per cent.

relatively highest level of Co in grain was found when the contamination took place at a time when the grain made up a substantial part of the plant. (Experiment VI does not come through in this and the following comparison because the time between spraying and harvest was so short that there had been no time for a redistribution of the activity within the plant.)

When the Cr/Fe ratio in grain was treated in a regression analysis, we got

$$(Cr/Fe)_{grain} = 0{\cdot}32 g{\cdot}cm^{-1} - 0{\cdot}17 \quad (r = 0{\cdot}9446), \qquad (3)$$

i.e. an equation quite similar to (2) and an indication of a preferential translocation of Fe to grain as compared with Cr. The translocation of Fe is, however, much less than that observed for Zn: the Fe/Zn ratio in grain was approx. 0·09 and nearly constant for experiments I–IV.

Figure 3 illustrates the greater translocation of Zn and Fe to the grain as compared with that of the other nuclides: the slopes are steeper for Co, Cr and Hg than for Zn and Fe.

Husks. There was no significant difference between the Zn, Fe, Cr, Co and Pb levels in husks from experiments III–VI. Hg, however, showed higher levels in these experiments than did the other nuclides. In experiments I and II the Zn and Co levels were higher than those of Fe and Cr as was the case with grain.

Straw. In experiments IV–VI there was no significant difference between the levels of the different nuclides in straw. In II and III the Zn and Co levels were significantly lower in straw than those of the other nuclides as could be expected from the relatively high Zn and Co levels in grain and husks.

Contamination models

CHAMBERLAIN[4] has introduced the term "normalized specific activity":

$$\text{N.S.A.} = \frac{\text{Activity kg}^{-1}\text{ dry weight of crop}}{\text{activity deposited day}^{-1}\text{ m}^{-2}\text{ of ground}}.$$

The term is analogous with the rate factor:

$$\varphi = \frac{\text{pCi·kg}^{-1}\text{ dry weight of crop}}{\text{mCi·km}^{-2}\text{·year}^{-1}}.$$

In the case of grain φ is based on the fall-out rate in the months prior to harvest and not on the whole year.[5] In the calculation of the N.S.A. for e.g. herbage, Chamberlain used mean of the daily fall-out rates for a two-month period, i.e. the month of cutting and the previous month.

Table 5 shows the N.S.A. values for the present and the 1967 experiment.[1] The conversion to natural conditions (0·45 kg grain m^{-2}) from experimental (1·44 kg m^{-2} in 1970 and 0·95 kg m^{-2} in 1967) is difficult. Not only the yield difference, but also the contamination method should be considered. On one hand the spraying procedure tends to give a higher retention on the leaves than raindrops, on the other hand we have dry fall-out under natural conditions which may add significantly to the activity levels of the herbage.[4,12,16]

In the calculation of N.S.A. for natural conditions we estimated that the natural initial

Table 5. Normalized specific activity (N.S.A.)[4] in barley grain under experimental and natural condition

Nuclide	N.S.A. experimental	N.S.A. natural	Remarks
^{210}Pb	1·2	~3	The activity data were obtained from
^{57}Co	5·0	~12	Tables 2 and 3. We used the mean levels
^{203}Hg	0·65	~1·5	found from expts. A and Σ/6.
^{51}Cr	1·2	~3	The period of contamination was ninety-
^{65}Zn	6·0	~15	seven days.
^{59}Fe	1·4	~3·5	(cf. also the text)
^{89}Sr	1·7	~2·5	The activity data are from reference (1).
^{134}Cs	6·1	~9·5	The period of contamination was sixty-
^{54}Mn	3·5	~5·5	eight days.
^{141}Ce	1·5	~2·5	(cf. also the text).
^{90}Sr	—	3·9	The rate factors found for 1962–1968[15]
^{137}Cs	—	7·5	were used in the calculations. Mn is the
^{54}Mn	—	8±3	mean of the values found in 1963 and 1964 (±1 SE).

uptake was 75 per cent of the artificial*. The comparison in Table 5 between the empirical N.S.A. values for ^{90}Sr, ^{137}Cs and ^{54}Mn, and $N.S.A._{nat.}$ based on the 1967 experiment shows that 75 per cent can be used as a rough estimate.

SELMER-OLSEN *et al.*[12] have measured the lead concentration in samples collected at different distances from a main road in Norway near Oslo. They found in 1969 0·03–0·05 mg Pb/l precipitation at a distance of 50–100 m from the road. During the growing season for barley the mean precipitation in Oslo[7] is ~ 330 mm. Hence the fallout rate becomes 0·08–0·13 mg Pb m^{-2} day^{-1}. In Table 5 the N.S.A. for natural conditions is estimated at 3, and this means that the expected level in barley grain becomes 0·24–0·40 mg Pb kg^{-1}. SELMER-OLSEN *et al.*[13] found 0·3–0·5 mg Pb kg^{-1} in barley grain grown 100–50 m from the road.

The fall-out rate of approx. 0·1 mg Pb m^{-2} day^{-1} is probably low for densely populated areas. Thus F. G. KLOKE *et al.*[9] found 3·4 mg Pb/kg barley grain in West Germany at locations distant from traffic.

High lead content might, however, also be a result of lead uptake through the roots from the soil. The importance of this kind of contamination with lead has not yet been completely clarified. The literature[6,9,10] gives contradictory information on this point. The present experiment, however, shows that direct contamination is likely to be an important route for lead in grain.

SUMMARY AND CONCLUSIONS

The initial retention in growing barley was related to the surface of the plants at the time of spraying, if the surface is expressed as the ratio between the dry weight of the plants and their height.

If the plants were contaminated throughout the growing season, the distribution of Pb, Hg, Cr and Fe followed nearly the same pattern within the plant, i.e. 10 per cent in grain, 30–35 per cent in husks and the remainder in straw. Co and Zn showed 40–40 per cent in grain, 25–30 per cent in husks and the rest in straw.

During the first part of the growing period the plants showed a rapid loss of activity. The field loss corresponded to a half-life of approx. two weeks. In the second part of the growing period, when the plants had nearly attained their final size and shape, the field loss was lower. In this period most nuclides showed a half-life of six seven-weeks. For a growing plant such as barley where the morphology changes rather rapidly, the field loss could either be described by means of a number of exponential equations each covering some part of the total growing period, or, if the whole period is considered, by means of a linear function, where the loss of activity from the foliage is proportional to the number of days from spraying until harvest.

The order for direct contamination of barley grain was: Zn > Co > Fe > Cr > Pb ~ Hg. Zn was very easily translocated within the plant contrary to Cr, Pb and Hg; Co and Fe took up an intermediate position.

The experiment showed that a substantial part of the lead content of grain might be explained by the direct contamination of crops by airborne lead.

It should be recognized that the conclusions reached from the present experiment may not be applicable to field conditions because the spraying procedure represents somewhat of an artifact. In the field the plants receive the activity in large volumes of highly diluted solutions, and this might result in a different initial retention as compared with the present uptake from a small volume of a relatively highly concentrated solution.

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*Although the crop density was lower in the 1967 than in the present experiment, we used the same initial uptake in both cases, because the differences in drying conditions as mentioned above might have increased the initial uptake in 1967 and thus resulted in an overestimate of the N.S.A. experimental.

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REFERENCES

1. AARKROG A. (1969) On the direct contamination of rye, barley, wheat and oats with ^{85}Sr, ^{134}Cs, ^{54}Mn and ^{141}Ce. *Radiation Botany* **9**, 357–366.
2. AARKROG A. (1971) Prediction models for strontium-90 and caesium-137 levels in the human food chain. *Health Phys.* **20**, 297–311.
3. CHADWICK R. C. and CHAMBERLAIN A. C. (1970) Field loss of radionuclides from grass. *Atmos. Environ.* **4**, 51–56.
4. CHAMBERLAIN A. C. (1970) Interception and retention of radioactive aerosols by vegetation. *Atmos. Environ.* **4**, 57–78.
5. COMAR C. L. (1969) Some thoughts about radioactive contamination of the environment from peace-time applications of atomic energy, pp. 91–96. *In, Environmental contamination by radioactive materials.* Proceedings of a seminar, Vienna, 1969. IAEA, Vienna.
6. TER HAAR G. (1970) Air as a source of lead in edible crops. *Environ. Sci. Technol.* **4**, 226–229.
7. HASL-239 (1971) *Fall-out program quarterly summary report.* Appendix p. A-237. Health and Safety Laboratory, New York, N.Y.
8. HULL C. R. (1960) Lead-210 and polonium-210 in grass. *Nature* **187**, 211–212.
9. KLOKE A. und LEH H. O. (1969) Verunreinigungen von Kulturpflanzen mit Blei aus Kraftfahrzeugabgasen, pp. 259–268. *In, Proceedings of the air pollution congress on plants and animals*, 1st, Wageningen, Netherlands, 1968. Centre for Agricultural Publishing and Documentation, Wageningen.
10. LAGERWERFF J. V. (1970) Air as a source of lead in edible crops. Comments. *Environ. Sci. Technol.* **4**, 230.
11. MOORBY J. and SQUIRE H. M. (1963) The loss of radioactive isotopes from the leaves of plants in dry conditions. *Radiation Botany* **3**, 163–167.
12. PERKINS R. W., THOMAS C. W. and NIELSEN J. M. (1965) Measurements of airborne radionuclides and determination of their physical characteristics, pp. 198–221. *In* A. W. KLEMENT (ed.), *Radioactive fall-out from nuclear weapons tests.* Proceedings of the 2nd Conference, Germantown, Pa., 1964. USAEC, Oak Ridge, Tenn.
13. SELMER-OLSEN A. R. and MYHRE A. (1970) En undersøkelse av blyinnholdet i plantemateriale langs Mosseveien. *Sci. Rept Agric. Coll. Norway* **49**, 1–7.
14. UNSCEAR (1962) *Report of the United Nations Scientific Committee on the Effects of Atomic Radiation*, UN, New York, 442 pp.
15. WILSON D. W., WARD G. M. and JOHNSON J. E. (1967) Fallout ^{137}Cs: Direct aerial transfer as an important source of foliar deposition. *Radiation Botany* **7**, 312–319.

Health Physics Pergamon Press 1975. Vol. 28 (May), pp. 557–562. Printed in Northern Ireland

RADIONUCLIDE LEVELS IN MATURE GRAIN RELATED TO RADIOSTRONTIUM CONTENT AND TIME OF DIRECT CONTAMINATION

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Abstract—Information on the relative behaviour of radioelements besides the well-known fallout nuclides ^{90}Sr and ^{137}Cs in cereal crops may be important in the assessment of monitoring programmes after unintentional releases of radioactivity to the terrestrial environment, be it airborne or transferred from the aquatic milieu in contaminated irrigation water.

In the present study a variety of radionuclides were administered as a fine spray to cereal crops at different stages of development. The direct contamination of cereals depends greatly on the stage of development of the plants. Certain nuclides, e.g. ^{90}Sr, ^{144}Ce and ^{106}Ru will hardly show up in the grain, if contamination takes place at the early stages of development. Nuclides such as ^{65}Zn, ^{55}Fe, ^{137}Cs, ^{60}Co and ^{54}Mn are, on the other hand, easily translocated to the seeds, and may therefore be present in cereal products even if crops have been contaminated several weeks before ear emergence.

The radionuclide levels in grain relative to radiostrontium follows with approximation the equation: $r = ae^{-bt}$, where r is the relative level and t is the time from ear emergence to contamination of the crops; a and b are constants which characterize the translocation of a nuclide relative to radiostrontium. In case of an environmental contamination of cereal crops by known (relative) amounts of ^{90}Sr and γ-emitting radionuclides it is possible to use the above equation to estimate the ^{90}Sr levels in the mature grain from γ-spectroscopical determination of the accompanying nuclides, thus avoiding laborious radiochemical analyses.

INTRODUCTION

TERRESTRIAL vegetation mainly takes up radionuclides by direct contamination. Studies of the radioactive fallout from nuclear weapons testing (UNSCEAR, 1958, 1962, 1964, 1966, 1969 and 1972) and experimental contamination (MIDDLETON, 1969; MIDDLETON and SQUIRE, 1961; AARKROG, 1969) of various crops have elucidated the contamination mechanisms for common fission products such as ^{137}Cs and ^{90}Sr.

Cesium-137 is translocated within the plants, i.e. this radionuclide will be present in the grain products even if the contamination has occurred at an early stage of development. Strontium-90 on the other hand shows relatively little internal transport in the plants, and if contamination of a crop takes place during the first part of the growing period, only minor amounts of ^{90}Sr will be found in the mature grain.

Beside fission products nuclear installations also release activation products, e.g. ^{55}Fe, ^{65}Zn, ^{54}Mn, $^{58,60}Co$, $^{124,125}Sb$, ^{134}Cs and ^{51}Cr. The dissemination of these products is mainly to the aquatic milieu, and radioecological studies of these radioelements have therefore been carried out only to a limited extent for the terrestrial environment.

As releases of activation products to the terrestrial milieu cannot be precluded in emergency situations or perhaps as a result of use of contaminated water for irrigation purposes, information on the behaviour of these radioelements is needed.

Since 1967 experiments with artificial, direct contamination of grain species, mainly barley, have been carried out at Risø. Until now fifteen elements have been studied, namely Sr, Cs, Ce, Ba, Sb and Ru representing fission products, Zn, Fe, Co, Mn, Na and Cr representing activation products, and finally three representatives of non-radioactive contamination: Cd, Hg and Pb.

The aim of the present investigation was to study the relative translocation of radioisotopes of Ru, Sr, Sb, Mn, Co, Cs and Fe in barley and

wheat plants and to propose a quantitative expression for the translocation ability of a given nuclide relative to radiostrontium. Information on the relative translocation of radionuclides is useful in the formulation of monitoring programmes in case of radioactive contamination of the terrestrial environment.

EXPERIMENTAL METHODS

The study was carried out in the summer of 1972 according to the principles used previously (AARKROG, 1969; AARKROG and LIPPERT, 1971). The experimental field was divided into 65 plots. Each of five experiments comprised six plots with barley, six with wheat and one with barley or wheat used as background control. Two experiments were contaminated before emergence of the ears, and three afterwards.

To avoid possible edge effects the experimental circular plot (1/8 m^2) was surrounded by a larger (1 × 1 m^2) plot where the same species were grown as in the experimental plot. Before the spraying we put a plastic ring round the experimental plot. This ring was left in the plot until harvest so that we could identify treated plants from untreated.

Each circular plot was sprayed with 10 ml of spraying solution and then with 10 ml of distilled water. The 10 ml of spraying solution contained on 1 May 1972: 8 μCi ^{85}Sr, 100 μCi ^{59}Fe, 6 μCi ^{54}Mn, 0.6 μCi ^{57}Co, 6 μCi ^{125}Sb, 2 μCi ^{134}Cs and 20 μCi ^{103}Ru. The carrier contentration was 10 ppm except for Fe and Mn where it was 25 ppm. The solution contained 0.1 M citric acid and 0.025 M oxalic acid/per l., and the pH was 1.6.

Before the measurements the plant material was dried at 80°C for 24 hr and divided into grain, husks and straw in a glove box. The radioactivity was determined on a 30 cm^3 Ge(Li) detector connected to a 1024-channel Hewlett–Packard analyser together with an H.P.-9810A calculator. The following photopeaks were used: ^{57}Co: 122 keV, ^{125}Sb: 427 keV, ^{103}Ru: 497 keV, ^{85}Sr: 514 keV, ^{134}Cs: 605 keV, ^{54}Mn: 835 keV and ^{59}Fe: 1095 keV. The activity levels in a sample were given as a percentage of a standard containing 10 ml of the spraying solution, i.e. all level were automatically corrected for radioactive decay, and the activity administered to a plot was 100% for all nuclides and experiments.

RESULTS AND DISCUSSION

To overcome the difficulties in the interpretation of the results which might arise from difficulties in growth of the crops, from variations in the administration of the spray, or from local variations in the experiment field, the nuclides were all related to ^{85}Sr, i.e. we determined the ratios: ^{103}Ru/^{85}Sr, ^{125}Sb/^{85}Sr, ^{54}Mn/^{85}Sr, ^{134}Cs/^{85}Sr and ^{59}Fe/^{85}Sr (in the following we shall omit the atomic weights).

Table 1 summarizes the experimental data. The yields in the experimental plots were approx 25% lower than under normal Danish agricultural conditions. Tables 2 and 3 show the level of the various nuclides in the mature crops. Figure 1 shows the radionuclide ratios in the different parts (grain, husks and straw) of mature wheat and barley related to the contamination time. If there had been no difference in the behaviour of the seven nuclides, all ratios would have fluctuated around unity, and the variations would all have been due to experimental errors. It is, however, evident from Fig. 1 that the experimental errors were not sufficient to explain the deviations of the curves from unity.

Table 1. Experimental data

Experiment No.	Spraying date	Height of crops (cm) Barley	Wheat	Dry-matter weight of herbage at spraying (g ± 1 S.E.) Barley	Wheat	Dry-matter weight of herbage at harvest (g ± 1 S.E.) Barley	Wheat	Grain yields of mature plants (g ± 1 S.E.) Barley	Wheat	Accumulated amount of precipitation since first spraying (mm)
I	6 June	31	38	16 ± 1	11 ± 0	75 ± 6	66 ± 7	37 ± 3	24 ± 3	0
II	21 June	56	57	35 ± 4	36 ± 1	96 ± 9	95 ± 4	48 ± 5	48 ± 5	37
III	5 July	78	81	55 ± 3	69 ± 22	92 ± 12	91 ± 4	46 ± 7	37 ± 1	39
IV	26 July	78	94	95 ± 0	79 ± 14	82 ± 10	98 ± 12	41 ± 5	39 ± 6	56
V	17 August	62	93	83 ± 8	88 ± 7	79 ± 2	81 ± 13	41 ± 0	35 ± 4	112
Harvest	28 August	—	—	—	—	—	—	—	—	114

The grain was sown on 4 April. The seeds came up around 20 April. The ears emerged between experiments II and III around 1 July. The experimental plot was 1/8 m^2.

Table 2. Per cent of activity in 10-ml spraying solution dosed on a plot in mature, grain, husks and straw (corrected for radioactive decay)

Barley	Exp.	^{57}Co	^{125}Sb	^{103}Ru	^{85}Sr	^{134}Cs	^{54}Mn	^{59}Fe
Grain	I	0.10 ± 0.04	0.011 ± 0.007	—	—	0.16 ± 0.06	0.080 ± 0.024	0.70 ± 0.27
	II	0.24 ± 0.04	0.075 ± 0.018	0.008 ± 0.002	0.012 ± 0.005	0.45 ± 0.07	0.21 ± 0.05	0.57 ± 0.08
	III	1.43 ± 0.24	0.92 ± 0.16	0.16 ± 0.02	0.14 ± 0.03	2.15 ± 0.32	0.47 ± 0.10	1.34 ± 0.22
	IV	1.48 ± 0.21	0.99 ± 0.16	0.51 ± 0.11	0.89 ± 0.14	1.14 ± 0.16	0.84 ± 0.12	1.00 ± 0.17
	V	1.18 ± 0.18	1.26 ± 0.20	1.18 ± 0.18	1.24 ± 0.20	1.68 ± 0.26	1.25 ± 0.20	1.04 ± 0.18
Husk	I	0.039 ± 0.012	0.031 ± 0.015	—	—	0.050 ± 0.015	0.031 ± 0.008	0.077 ± 0.025
	II	0.12 ± 0.03	0.18 ± 0.04	0.10 ± 0.03	0.074 ± 0.019	0.17 ± 0.03	0.10 ± 0.02	0.14 ± 0.04
	III	0.53 ± 0.07	1.19 ± 0.16	1.46 ± 0.18	0.84 ± 0.13	0.81 ± 0.13	0.68 ± 0.09	1.13 ± 0.18
	IV	2.39 ± 0.32	2.52 ± 0.25	2.59 ± 0.23	2.69 ± 0.31	1.46 ± 0.10	2.94 ± 0.31	3.28 ± 0.40
	V	9.30 ± 0.54	10.1 ± 0.6	9.57 ± 0.58	9.91 ± 0.77	9.18 ± 0.88	9.08 ± 0.64	10.1 ± 0.5
Straw	I	0.64 ± 0.20	0.86 ± 0.31	1.18 ± 0.44	0.91 ± 0.36	0.85 ± 0.23	0.96 ± 0.32	1.08 ± 0.34
	II	1.85 ± 0.27	1.78 ± 0.27	2.79 ± 0.38	3.81 ± 0.55	2.01 ± 0.28	2.73 ± 0.42	3.05 ± 0.52
	III	2.07 ± 0.15	2.42 ± 0.06	3.51 ± 0.16	4.11 ± 0.16	2.67 ± 0.16	3.06 ± 0.14	3.39 ± 0.14
	IV	5.04 ± 0.23	5.25 ± 0.71	4.41 ± 0.50	6.42 ± 0.58	3.36 ± 0.11	6.87 ± 0.59	5.37 ± 0.28
	V	10.5 ± 0.2	9.28 ± 0.29	9.22 ± 0.24	10.4 ± 0.4	9.75 ± 0.23	10.3 ± 0.2	9.61 ± 0.16

The error term is the S.E. of the mean of triple determinations.

Variation between species

The Fe/Sr ratio was generally higher ($P > 97.5\%$) in grain of wheat than of barley, while the ratio in husks and straw did not differ significantly for the two species.

In the case of the Cs/Sr ratio we found a significant ($P > 99\%$) interaction between species and experiments for grain and a significant ($P > 99.5\%$) second-order interaction between species, experiments and plant parts, but only for husks and straw. It was thus not possible generally to state whether Cs was translocated more extensively in barley than in wheat relative to Sr, because in some experiments some plant parts of wheat showed the highest Cs/Sr ratios, in others those of barley did.

The Mn/Sr ratio was significantly ($P > 99\%$) greater in wheat than in barley grain, while the ratio in straw was lower for wheat than for barley ($P > 99.95\%$). Husks did not differ for the two species. It was thus evident that, relative to Sr, Mn was translocated to a greater extent in the wheat plant than in barley.

The Co/Sr ratio showed the same pattern between wheat and barley as Mn/Sr, but the difference between species was not significant in this case.

The distribution of Sb/Sr and Ru/Sr within wheat and barley resembled the pattern found for Cs/Sr. The second-order interactions in the anovas were significant, i.e. the relative lapse of the curves (for a given ratio) for the plant parts as a function of experiment was different for wheat and barley (cf. Fig. 1).

Hence we may conclude that all the six ratios (Fe/Sr, Cs/Sr, Co/Sr, Mn/Sr, Sb/Sr and Ru/Sr) were different in wheat and barley. However, only in the case of Mn/Sr did we find a systematically greater translocation for wheat than for barley.

Table 3. Per cent activity in the 10-ml spraying solution dosed on a plot in mature, grain, husks and straw (corrected for radioactive decay)

Wheat	Exp.	^{57}Co	^{125}Sb	^{103}Ru	^{85}Sr	^{134}Cs	^{54}Mn	^{59}Fe
Grain	I	0.38 ± 0.08	0.004 ± 0.002	—	—	0.20 ± 0.03	0.13 ± 0.03	0.65 ± 0.13
	II	0.84 ± 0.03	0.034 ± 0.002	0.004 ± 0.004	0.008 ± 0.004	0.81 ± 0.05	0.41 ± 0.09	1.05 ± 0.05
	III	2.24 ± 0.14	0.34 ± 0.04	0.052 ± 0.039	0.23 ± 0.02	2.58 ± 0.23	1.01 ± 0.08	2.59 ± 0.15
	IV	4.90 ± 1.94	1.48 ± 0.38	0.30 ± 0.10	0.59 ± 0.25	2.51 ± 0.97	1.85 ± 0.49	2.58 ± 0.76
	V	0.47 ± 0.05	0.29 ± 0.01	0.41 ± 0.07	0.98 ± 0.12	1.05 ± 0.10	0.82 ± 0.11	0.95 ± 0.09
Husk	I	0.14 ± 0.01	0.021 ± 0.001	—	—	0.098 ± 0.009	0.059 ± 0.006	0.14 ± 0.01
	II	0.22 ± 0.02	0.21 ± 0.02	0.066 ± 0.024	0.021 ± 0.006	0.36 ± 0.04	0.15 ± 0.03	0.17 ± 0.02
	III	0.67 ± 0.09	1.56 ± 0.12	1.28 ± 0.15	0.98 ± 0.08	1.47 ± 0.14	0.70 ± 0.10	1.00 ± 0.14
	IV	2.18 ± 0.78	3.44 ± 1.32	3.02 ± 1.06	3.54 ± 1.34	2.53 ± 0.84	3.06 ± 1.14	3.03 ± 1.05
	V	9.21 ± 0.45	10.7 ± 0.5	10.4 ± 0.5	10.4 ± 0.6	9.95 ± 0.63	10.1 ± 0.6	10.9 ± 0.5
Straw	I	0.48 ± 0.04	1.53 ± 0.17	1.57 ± 0.25	1.08 ± 0.14	1.39 ± 0.19	0.75 ± 0.13	1.30 ± 0.18
	II	1.38 ± 0.03	3.43 ± 0.08	3.69 ± 0.15	4.56 ± 0.07	3.42 ± 0.09	2.50 ± 0.19	3.46 ± 0.08
	III	1.61 ± 0.12	4.84 ± 0.49	4.61 ± 0.59	5.44 ± 0.48	4.30 ± 0.50	2.34 ± 0.24	4.12 ± 0.55
	IV	3.06 ± 0.42	4.89 ± 1.47	7.00 ± 2.29	7.99 ± 2.07	4.58 ± 0.87	5.68 ± 1.69	7.19 ± 1.54
	V	7.40 ± 1.00	7.09 ± 0.83	6.83 ± 0.87	7.29 ± 1.11	7.14 ± 0.76	7.16 ± 0.86	7.38 ± 0.96

The error term is the S.E. of the mean of triple determinations.

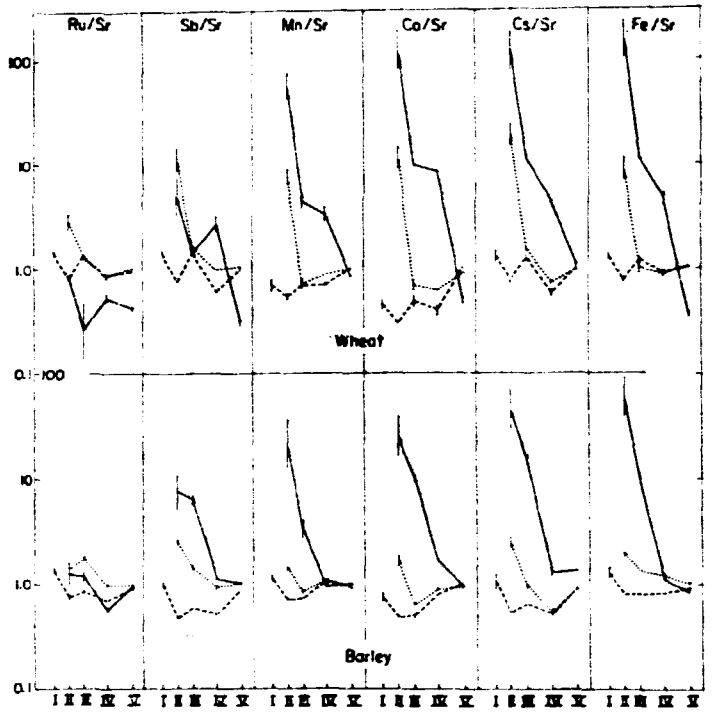

FIG. 1. Radionuclide ratios in grain (——), husks (· · · · · ·) and straw (- - - - - -) of mature wheat and barley related to contamination time (cf. Table 1). The vertical bars indicate ±1 S.E. of the mean of triple determinations. Experiment I was omitted for grain and husks because only one Sr determination was available for these plant parts in that experiment. The abscissa and the ordinate indicate the experiments I–V and the radionuclide ratios r, respectively.

Anovas on the Ru/Cs, Sb/Cs, Mn/Cs and Co/Cs ratios showed that compared with Cs, Ru and Sb were translocated to a greater extent in barley than in wheat, whereas the opposite was the case for Mn and Co.

Variation between plant parts

In most cases the ratios (cf. Fig. 1) were greater in grain than in husks, which in their turn were greater than in straw. The lapses of the curves were, however, very different for the three plant parts. The grain ratios showed the steepest (negative) slope. The ratios in straw did not generally show any marked slopes, and when they did, the slopes were positive rather than negative. Ratios of Sb/Sr in wheat and Ru/Sr in both species did not follow the pattern above. For these ratios we found no common tendency as we did for the other ratios.

We may conclude that Fe, Cs, Co, Mn and Sb (in barley) showed a translocation relative to Sr from straw to husks and from husks to grain. Ruthenium was not translocated relative to Sr.

Variation between experiments

Figure 1 shows that all ratios in grain and husks, except Ru/Sr, showed higher levels in the first experiments than in the last.

We may again conclude that Fe, Cs, Co, Mn and Sb were all translocated less to the upper parts of the parts of the plant, relative to Sr, as time went by. In agreement with this the ratios in straw tended to increase with time. Since the activity reservoir in straw generally was greater than in the other parts of the plant, it was not possible to observe any consistent influence of the translocation to the grain on the ratios in straw.

Variation between nuclide ratios

In Fig. 1 the ratios have been arranged according to the steepness of the slopes for grain. The slopes were used as an expression for the translocation ability—the steeper the slope, the greater the translocation rate.

The curves for the ratios in grain approximated straight lines, i.e. the ratios (r) varied roughly exponentially with time.

By regression the data were fitted to the equation:

$$r = ae^{-bt}, \qquad (1)$$

where the independent variable t is the time in days from ear emergence to contamination of the plot; t is negative if the spraying was before emergence of the ears, and positive if it took place afterwards; it is a measure of the stage of development of the crops; a is a constant which states the nuclide ratio if the contamination took place at the time of ear emergence; b is the relative translocation rate factor. Table 2 shows a summary of the constants for the regression lines for the different ratios.

Although we observed differences between wheat and barley in the internal transport of the nuclides, we shall consider such differences unimportant in the present context and make no distinction between the species in the regression analysis. In most cases we found significant deviation of the data from the regression line. This could mean either that the logarithms of the ratios were curvilinear functions of t or that there was a large amount of random heterogeneity around the regression line (or perhaps a mixture of the two). We supposed heterogeneity was responsible for the significant deviation, and we tested the significance of linear regression against the deviation from regression, and found significant linear regression for all ratios (except Ru/Sr). The radionuclides according to the translocation rate factor b can be arranged as follows:

$$\text{Fe} \geqslant \text{Cs} \sim \text{Co} \sim \text{Mn} > \text{Sb} > \text{Sr},$$

i.e. Sb was translocated less rapidly than Mn, Co and Cs which was probably translocated more slowly than Fe. All five nuclides were translocated more rapidly than Sr.

It is interesting to notice that for the ratio in equation (1) equal to unity, i.e. at the contamination time where no further translocation occurred between spraying and harvest, t averaged 40 ± 2 days (±1 S.E.) for the five ratios. This is in agreement with the time when the crops have matured (PEDERSEN, 1963). After maturity we assumed that no active transport of nuclides took place within the plant.

Experiments in earlier years

In 1967, ^{141}Ce, ^{85}Sr, ^{134}Cs and ^{54}Mn were studied (AARKROG, 1969) in wheat, barley, oats and rye. If regression equation (1) is applied to these data, we get the values for a and b shown in Table 2. The last experiment in 1967 was not included in the data as the spraying took place the day before harvest, i.e. 2–3 weeks after maturity. The values are in reasonable agreement with the 1972 data in Table 4 considering that species as well as contamination times and growing conditions were different in the two years.

In the studies in 1970 (AARKROG and LIPPERT, 1971) and 1971 (AARKROG, 1972), Sr was not included among the nuclides examined. It was therefore necessary to use other reference nuclides, and the determination of the radionuclide ratios for these years thus becomes more uncertain. Table 4 shows the estimated constants to be substituted into equation (1).

We may now arrange the nuclides examined according to their translocation ability as follows:

$$\text{Zn} \geqslant \text{Fe} \geqslant \text{Cs} \geqslant \text{Co} \geqslant \text{Mn} \geqslant \text{Na} \geqslant \text{Cd} \geqslant \text{Sb} \geqslant \text{Cr} \geqslant \text{Sr} \sim \text{Ce} \sim \text{Ru} \geqslant \text{Ba} \sim \text{Hg} \sim \text{Pb}.$$

Like Ru, Ba, Hg, Pb and Ce showed no translocation relative to Sr.

The behaviour of certain nuclides may vary with their chemical state, e.g. the oxidation step. The internal transport may also be influenced by complexing agents. We shall therefore consider the above mentioned order of translocation only as a rough guide.

Practical applications

It is well known that ^{90}Sr belongs to the group of most hazardous radionuclides. Its determination in environmental samples is laborious because γ-spectroscopy cannot be applied. In the case of emergency monitoring of contamination of cereal crops it may be possible to avoid or at least to reduce substantially the number of ^{90}Sr

Table 4. The constants in the regression line: nuclide ratio = ae^{-bt} (data from 1967, 1970, 1971, and 1972)

Ratio	Experimental year	Species	a	$b \pm 1$ S.E.	Remarks	Reference
Fe/Sr	1972	wheat barley	27.5	0.087 ± 0.006		
Cs/Sr	1972	wheat barley	22.4	0.071 ± 0.005		
Co/Sr	1972	wheat barley	20.2	0.072 ± 0.006		
Mn/Sr	1972	wheat barley	11.1	0.060 ± 0.006		
Sb/Sr	1972	wheat barley	3.7	0.038 ± 0.005		
Cs/Sr	1967	rye barley wheat oats	34	0.08	The last experiment in 1967 was excluded from the calculation.	Aarkrog (1969)
Mn/Sr	1967	rye barley wheat oats	16	0.07		
Zn/Sr	1970	barley	100	0.10	Estimated from the Zn/Fe, Zn/Co, Fe/Cr, Co/Cr ratios in 1970 and the Fe/Sr and Co/Sr ratios from 1972	Aarkrog and Lippert (1971)
Cr/Sr	1970	barley	3	0.02		
Na/Sr	1971	barley	8	0.03	Estimated from the Cs/Na, Cs/Cd, Na/Sb, and Cd/Sb ratios in 1971 and the Cs/Sr and Sb/Sr ratios in 1972	Aarkrog (1971)
Cd/Sr	1971	barley	4	0.05		

The date of ear emergence, $t = 0$, was fixed at 1 July for all years.

analyses by applying values in Table 4 an equation (1).

Let us suppose we know the relative or absolute amounts of the different nuclides including ^{90}Sr deposited from an accident. Let us further assume that by γ-spectroscopy we have determined the γ-emitters in the mature crops and that their half-lives are long (>3/4 yr) as compared with the biological half-life in the crop which is 2–3 weeks (Aarkrog, 1969, 1972; Aarkrog and Lippert, 1971) for most nuclides. From information on the time of contamination it is then possible from equation (1) and Table 4 to estimate the ^{90}Sr level arising from the accident in the mature grain. The estimate will of course be improved if more than one γ-emitter has been used in calculation.

CONCLUSION

The internal transport of ^{103}Ru, ^{125}Sb, ^{54}Mn, ^{57}Co, ^{137}Cs and ^{59}Fe relative to ^{85}Sr differed in wheat and barley. The picture was in most cases obscured by significant second- and first-order interactions. It was, however, evident that relative to Sr, Mn was translocated to a greater extent in wheat than in barley.

The nuclide ratios r: Fe/Sr, Cs/Sr, etc. in mature grain could with approximation be related to the time (t) of contamination by an exponential expression: $r = ae^{-bt}$, a and b being constants describing the translocation ability of a nuclide relative to Sr.

According to the a and b values the different nuclides were ordered after translocation ability:

$$\text{Fe} \geqslant \text{Cs} \sim \text{Co} \sim \text{Mn} > \text{Sb} > \text{Sr} \sim \text{Ru}.$$

After an accidental release of known amounts of ^{90}Sr and γ-emitting radionuclides to the terrestrial environment, the lengthy radiochemical determination of radiostrontium in cereal crops could be avoided. If one knows the relative amounts of the released activities and makes a γ-spectroscopical analysis on the mature grain, it is by means of the above expression possible to estimate the ^{90}Sr level in mature cereal grain.

REFERENCES

Aarkrog A., 1969, *Radiat. Bot.* **9**, 357.

Aarkrog A., 1972, Risø Rep. No. 256, pp. 163–175.

Aarkrog A. and Lippert J., 1971, *Radiat. Bot.* **11**, 463.

Middleton L. J., 1959, *Int. J. Radiat. Biol.* **1**, 387.

Middleton L. J. and Squire H. M., 1961, A.R.C. *Radiobiol. Lab. Rept.* **5**, 50.

Pedersen A., 1963, *Landbrugets Plantekultur* II. Royal Veterinary and Agricultural University, Copenhagen 1963, 38 (in Danish).

UNSCEAR, 1958, 1962, 1964, 1966, 1969 and 1972, Report of the United Nations Scientific Committee on the Effects of Atomic Radiation.

0017-9078/78/0901-0489/$02.00/0
Health Physics Vol. 35 (September) pp. 489–491
Pergamon Press Ltd. 1978. Printed in Great Britain
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Variation of Direct Plutonium Contamination in Danish Cereal Grains

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Introduction

DURING the early 1960's when the fallout rate from nuclear weapons testing was reduced, grain products were the main contributors of radionuclides, such as ^{90}Sr, ^{137}Cs and ^{54}Mn, in the Danish diet (Aa64). In a 1972 study of plutonium in the New York diet, Bennett (Be74) found wheat products in the diet to be among the most important foodstuffs with respect to the intake of plutonium. It was thus obvious to study the plutonium concentrations in Danish cereal grain.

The root-uptake of plutonium, i.e. indirect contamination, has been studied for cereals such as wheat (Sc75), barley (Sc75; Wi74), and oats (Cu71; Gi71). Root-uptake, however, only plays a minor role for the plutonium concentrations in cereals that originate from newly deposited radioactive debris. In this case direct contamination is the most important way of exposure. Nevertheless, very few results have been published on direct Pu-contamination of cereals; and only wheat has been examined to date (Be74; Fr75; Ad75).

Materials and Methods

The present study was carried out on grain samples of wheat, rye, barley, and oats collected in 1963 and 1965 from eleven Danish State Farms distributed over the country (Aa64). These 2 yr were selected to obtain a set of sample populations which, on the one side, differed significantly with respect to fallout rate (approx a factor of five) and, on the other, showed sufficiently high concentrations to ensure a reliable plutonium determination. Plutonium was analyzed on 1–5 g grain ash by the classical anion-exchange method (Ta71) and α-counted for 3 days on a Si-detector; the standard deviation resulting from counting was about 15%.

Results

The results were treated by a three-way analysis of variance (anova) (Ac77), which showed a highly significant variation ($P > 99.9\%$) between years and among species (Fig. 1). The variation among locations was not significant ($P \sim 85\%$), but showed the same pattern as observed for other radionuclides found in Danish grain (Aa64), i.e. the western, high-precipitation locations (760 mm yr^{-1}) showed higher levels than the eastern, low-precipitation stations (620 mm yr^{-1}). There were no significant interactions among years, species and locations. While the yearly variation of plutonium in grain followed that observed for ^{90}Sr and ^{137}Cs, the variation among species differed markedly. For direct contamination of cereals (Aa64) with ^{90}Sr, ^{137}Cs and ^{54}Mn, the general order of the concentrations of these nuclides has been: rye > barley > wheat ~ oats, but for plutonium the order was: barley > oats ~ rye > wheat, and the interspecific variance was significantly greater than that observed for other radionuclides.

The plutonium concentration in Danish wheat was approx 30% higher than that observed in US wheat in 1963 (Be74); but the Pu/Sr and Pu/Cs ratios were comparable in the two studies.

Analysis of variance of the ratios between plutonium and ^{90}Sr, ^{137}Cs and ^{54}Mn, respectively, in Danish grain showed that these ratios were significantly higher in barley than in wheat. As

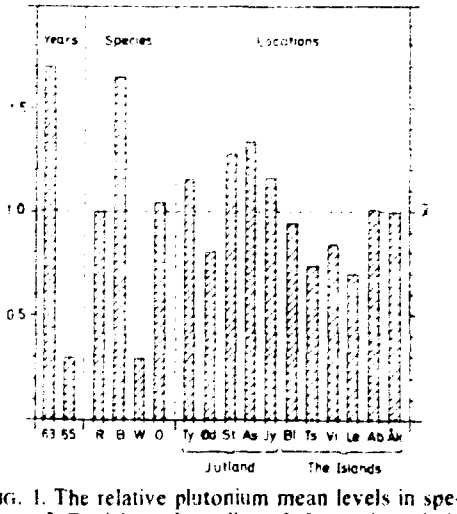

FIG. 1. The relative plutonium mean levels in species of Danish grain collected from the whole country in 1963 and 1965 calculated from the analysis of variance. The plutonium mean levels in the figure were dimensionless as they were obtained by division of the various group means with the grand mean: $\bar{x} = 0.66$ pCi $^{239,240}Pu$ kg^{-1}.

compared with the nuclide ratios in nuclear debris (Table 1), rye contained on the average 6% of the plutonium expected had the plutonium shown an uptake similar to that of the other nuclides; barley contained 15%, wheat 3% and oats 9%. From these percentages, and from the calculated transfer factors for ^{90}Sr, ^{137}Cs and ^{54}Mn (Aa77: Aa 64) the transfer factors for plutonium can be estimated assuming the root-uptake of plutonium to be negligible: rye: 2, barley: 4, wheat: 0.7 and oats: 3 pCi Pu $kg^{-1} \cdot yr$ per mCi Pu km^{-2}.

Table 1. Radionuclide mean ratios in Danish grain and fall-out from May to August 1963 and 1965. (All ratios were multiplied by 10^3)

Sample	$^{239,240}Pu/^{90}Sr$	$^{239,240}Pu/^{137}Cs$	$^{239,240}Pu/^{54}Mn$ 1963	$^{239,240}Pu/^{54}Mn$ 1965
Rye	18 ± 2	7 ± 1	6 ± 2	28 ± 6
Barley	45 ± 3	18 ± 1	9 ± 1	84 ± 11
Wheat	7 ± 1	3.3 ± 0.4	1.9 ± 0.4	13 ± 3
Oats	28 ± 2	13 ± 1	7 ± 1	29 ± 8
Nuclear debris (Ca64; Aa64)	229	143	75	575

The error terms are 1 S.E.

Discussion

The observed variation of plutonium contamination in Danish cereal grain may be ascribed to the morphology of the grain species rather than to physiological peculiarities. In the case of wheat, the mutual protection of the grains in the relatively compact ear and the absence of awns in Danish wheat varieties reduce the direct contamination by insoluble particulate debris, such as plutonium fallout, and may furthermore facilitate the removal of adsorbed particles. As to barley, the inner glumes and the long awns may enhance the capture and the retention of particulate debris. Rye also has awns but the grain is bare as in wheat; oats on the other hand have no awns but do have inner glumes. This may explain why rye and oats lie between barley and wheat as to susceptibility to direct plutonium contamination.

To determine the role washing played in the concentrations, two samples of wheat grain and two of barley from 1963 were washed (150 g grain were stirred mechanically for 30 min in 1 l. of 20°C distilled water) and the Pu-ratios between washed and unwashed grain were determined. For barley, the mean ratio was 0.73 ± 0.02 (1 S.E.) and for wheat 0.46 ± 0.02, showing that the barley grain in fact retained its plutonium more efficiently than the wheat grain.

The consumption of rye bread made from whole-grain flour in particular is the main reason why Danish cereal products have been especially of interest to radioactive contamination. The present results thus called for an examination of the plutonium concentrations in bread. Rye bread contained 0.22 ± 0.05 (1 S.E.) of the Pu found in rye grain (by fresh weight). For ^{90}Sr, ^{137}Cs and ^{54}Mn, the corresponding mean was 0.69 ± 0.04. Thus approximately two-thirds of the plutonium had been removed from the rye during the manufacture of the bread. In the case of plutonium in white bread, the bread to wheat ratio was 0.09. This ratio was not significantly different from that observed for ^{90}Sr (0.11), or for ^{54}Mn (0.08), but was lower than that for ^{137}Cs (0.25), because ^{137}Cs is translocated more easily than the other nuclides. This shows that plutonium as well as ^{90}Sr and ^{54}Mn adhere mainly to the bran: approx 10% of these nuclides are absorbed in the depths of the grain and are not influenced by any washing. The difference between the plutonium concentrations in rye bread was not as pronounced as the plutonium levels in rye and wheat might have suggested.

Conclusion

The total deposition of $^{239,240}Pu$ in Denmark at the end of 1975 was 1.7 mCi km^{-2} (Aa77). From this figure, and from the above estimated transfer factors and bread/grain ratios, the infinite, time-integrated plutonium levels in Danish rye bread and white bread were estimated at 0.75 and 0.1 pCi Pu kg^{-1} yr, respectively. As the consumption rates of the two types of bread were approx 40 and 60 kg yr^{-1} per capita, the plutonium intake from the consumption of bread was 36 pCi per capita. The uptake of plutonium from the gastrointestinal tract was assessed at 3×10^{-5} (Un77); hence the wholebody content originating from Danish bread consumption was 1 fCi $^{239,240}Pu$. From the latest UNSCEAR publication (Un77) the total wholebody plutonium content in Danish individuals was estimated at 1.5 pCi. It is thus evident that, although Danish cereal products contained relatively high plutonium concentrations, their contribution to the wholebody content was not very significant.

Nearly all plutonium in the human body is derived from the inhalation pathway, and a situation where direct plutonium contamination of cereal grain is the critical pathway is unlikely.

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References

Aa64 Aarkrog A. and Lippert J., 1964–66, Environmental Radioactivity in Denmark 1963–65, Risø Report Nos 85, 107, 130.

Aa77 Aarkrog A. and Lippert J., 1977, Environmental Radioactivity in Denmark 1976, Risø Report No. 361.

Ad75 Adriano D. C., McLeod K. W., Pinder J. E. III, Smith M. E., Boni A. L., Coney J. C. and Horton J. H., 1975, Field Study to Determine Plutonium Contents of Wheat and Soil in a Warm Humid Area, Ann. Meeting Am. Soc. of Agronomy, DP-MS-75-40.

Be74 Bennett B. G., 1974, Fallout ^{239,240}Pu in Diet, USERDA-Report, HASL-286, I, pp. 36–52.

Ca64 Cambray R. S., Fisher E. M. R., Spicer G. S., Wallace C. G., Webber T. J., Brooks W. L. and Peirson D. H., 1964–66, Radioactive Fallout in Air and Rain, Harwell Reports, AERE-R Nos 4687, 4997, 5260.

Cu71 Cummings S. L. and Bankert L., 1971, Uptake of ^{144}Ce, ^{147}Pm, and ^{239}Pu by Oat Plants from Soils, Radiol. Health Data Report 12, pp. 83–85.

Fr75 Frindik O. and Diehl J. F., 1975, "The Plutonium Alpha Activity of Foodstuffs", (in German) *Dt. Lebensmitt Rdsch.* 71, 100–104.

Gn71 Gneusheva G. I., 1971, "Uptake of Plutonium-239 into Agricultural Plants from Soil", *Biol. Nauki* 14, 60–63.

Sc75 Schulz R. K., Tompkins G. A. and Babcock K. L., 1975, Uptake of Plutonium and Americium by Plants from Soils, Berkeley, UCB-34, pp. 211–2.

Ta71 Talvite N. A., 1971, "Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange", *Analyt. Chem.* 43, 1827.

Un77 UNSCEAR, 1977, Sources and Effects of Ionizing Radiation (New York: United Nations).

Wi74 Wildung R. E. and Garland T. R., 1974, "Influence of Soil Plutonium Concentration of Plutonium Uptake and Distribution in Shoots and Roots of Barley", *J. agr. Fd. Chem.* 22, 836–838.

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Health Physics Pergamon Press 1971. Vol. 20 (March), pp. 297–311. Printed in Northern Ireland

PREDICTION MODELS FOR STRONTIUM-90 AND CAESIUM-137 LEVELS IN THE HUMAN FOOD CHAIN

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Abstract—Milk, rye, barley, wheat, oats, potatoes, cabbage, carrots, beef, pork and total diet have been collected all over Denmark since the 1961–62 test series and analysed for ^{90}Sr and ^{137}Cs. Along with the performance of diet measurements, precipitation, soil and air samples were collected and analysed for ^{90}Sr. The diet levels were related to the fall-out data by a least squares analysis. It was not essential whether concentrations in air (or precipitation) or deposited activity were used in the "rate" and "lag" terms, although the deposition data tended to give the best models. Strontium-90 determinations in human bone and whole-body ^{137}Cs measurements were performed for the same period as the diet measurements. Quantitative models of the transfer of ^{90}Sr and ^{137}Cs to man were calculated in analogy with the equations used for diet OR $_{bone/diet}$ was calculated from the soil factors in the equations. The prediction models were transformed to exponential equations, which served for the calculation of the dose commitments to the Danish population.

INTRODUCTION

THE PRIMARY purpose of radioactivity measurements in the human food chain is to evaluate whether the radionucleides are present in concentrations that might be harmful to man. A secondary aim, which ultimately serves the primary end, is to collect information on the relation between the activity levels. From such surveys or experimental investigations it is possible to set up quantitative expressions which might serve as prediction models for future contamination levels.

The classic equation[1] for these prediction models was that proposed in 1958 for the yearly ^{90}Sr mean levels in milk, on the assumption that the ^{90}Sr in milk comes from:

(a) direct deposits on leaves, assumed to be proportional to the fall-out deposit in a one-year period, the fall-out rate being (F_r in mCi/km^2/yr);

(b) uptake through the roots of vegetation, assumed to be proportional to the accumulated deposit in soil (F_s in mCi/km^2).

The average ^{90}Sr level in milk, C_M, was found from

$$C_M = aF_r + bF_s, \qquad (1)$$

where a and b were proportionality constants, often described as the "rate" and "soil" factors. This classic equation was later improved by the introduction of a third term, the so-called "lag" rate factor,[2] which took into account the influence of fall-out in the preceding year on the contamination of pastures and stored food consumed by cattle:

$$C_M = aF_r + cF_1 + bF_s \qquad (2)$$

where F_1 is the deposition during e.g. the last 6 months of the preceding year and c a proportionality constant, the "lag" rate factor.

During the 5 yr period 1963–1967 the predominant source of long-lived fall-out was the debris from the 1961–1962 test series. During this period only small amounts of ^{90}Sr and ^{137}Cs originated from later tests.[3] Consequently it has been possible to follow the activity levels in the food chain, practically undisturbed by fresh amounts of fall-out, for a longer period than possible at any time since nuclear weapon testing began.

As during this 5 yr period the fall-out rate followed an exponential decay with reasonable approximation, it has been the aim to describe the various levels in the human food chain by exponential equations for these years.

The purpose was furthermore to obtain more reliable estimates of the soil factors in equations (1) and (2) than hitherto and especially to

investigate whether ^{137}Cs revealed any soil factors during the increase in importance of indirect contamination (soil uptake).

MATERIAL AND METHODS

In all of Denmark and the Faroes samples of air, precipitation, soil, the most important diet components (milk, cereals, vegetables, meat), total diet, and human tissue have been collected since 1962. The samples have been analysed for ^{90}Sr and in most cases also for ^{137}Cs. The results have been published currently in annual Risö Reports.[6]

Strontium-90 was determined by the conventional radiochemical analysis with fuming nitric acid separations,[6,8] and ^{137}Cs was measured by γ-spectroscopy on NaI crystals, mostly on fresh or dried samples.

The data were fitted to equation models (1) or (2) by the least squares analysis. For air and precipitation the natural logarithm of the activity levels was treated by regression analysis, which yielded a parameter estimate for the regression equation: $y = \alpha + \beta(t - \bar{t})$, where y was the mean of the natural logarithm of the activity levels, t the time in years since 1 January, 1963, $\bar{t}$ the mean of the t-values and α and β constants. In the tests for significance a variation was considered probably significant (i) for a probability level $p = 0.05$, significant (ii) for $p = 0.01$ and highly significant (iii) for $p = 0.001$.

RESULTS AND DISCUSSION

Air and precipitation

Figure 1 shows the annual mean levels of ^{90}Sr in air collected at two stations at Risö and in precipitation collected at ten state experimental farms all over Denmark. The regression analysis showed that it was probably significant that the air levels followed an exponential decay:

$$(\text{pCi } ^{90}Sr/10^3 \text{ m}^3 \text{ air}) = 67.5\, e^{-0.9t}, \quad (3)$$

and that it was highly significant that the ^{90}Sr level in precipitation decayed exponentially:

$$(\text{pCi } ^{90}Sr/\text{l. precipitation}) = 44.7\, e^{-0.70t} \quad (4)$$

and

$$(\text{mCi } ^{90}Sr/\text{km}^2) = 25.3\, e^{-0.71t}. \quad (5)$$

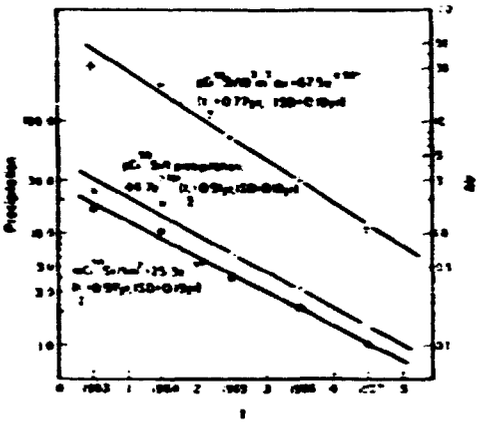

FIG. 1. Yearly mean values of ^{90}Sr in air and precipitation collected in Denmark compared with the regression lines (3), (4) and (5).

The annual mean levels were referred to the middle of the year, and to get the mean level for 1963, t was given the value 0.5; for 1964 $t = 1.5$, etc.

The effective half-lives in precipitation (0.91 yr and 0.97 yr) were significantly longer than those in air (0.77 yr). As far as the fall-out rate (5) was concerned, this was partly due to the fact that the amount of precipitation happened to be increasing throughout the period, which resulted in a lower decay rate of the mCi ^{90}Sr/km^2 figures. Tests were performed to determine whether the decay rate in precipitation differed at the different stations; that was not the case. However, it could not be ruled out that local conditions were the cause of the shorter half-life observed in the air samples.

The ^{90}Sr inventory in the stratosphere[7] decayed prior to June 1967 with a half-life of 10 months (0.83 yr). This is in accordance with the effective half-lives calculated above.

Since the end of 1967 the ^{90}Sr levels in fall-out have not decayed exponentially because the fall-out from the Chinese thermonuclear tests began to contribute significantly to the environmental levels. It has thus not been possible to use the fall-out data for exponential expressions for a period of more than 5 yr.

Soil

The ^{90}Sr level in the soil (the accumulated fall-out) depends on the fall-out rate and the

decay of the ^{90}Sr already deposited. From the solution of a differential equation the following expression was found:

$$A_t = \frac{\lambda_1}{\lambda_2 - \lambda_1} C_0(e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_0 e^{-\lambda_2 t}, \quad (6)$$

where A_0 is the accumulated fall-out in the soil (mCi $^{90}Sr/km^2$) on 1 January, 1963,
A_t the accumulated fall-out t yr later,
C_0 the amount of ^{90}Sr still present by 1 January 1963 as potential fall-out from the atmosphere, expressed in mCi $^{90}Sr/km^2$,
λ_1 the decay constant for ^{90}Sr in precipitation, and
λ_2 the decay constant for ^{90}Sr in the soil.

A_0 and C_0 were calculated from the soil measurements performed at ten Danish locations since 1961.[4] λ_1 was obtained from equation (5), and λ_2 was assumed to be equal to the physical decay constant for ^{90}Sr. Equation (6) could now be rewritten as

$$A_t = 65.0\, e^{-0.025t} - 36.3\, e^{-0.71t}. \quad (7)$$

If equation (7) is differentiated, and $dA_t/dt = 0$, the time, t_{max}, at which the accumulated fall-out reached its maximum, may be calculated. At $t = 4.05$ yr the soil maximum occurred, i.e. in January 1967. This is an agreement with the estimate of HARDY *et al.*,[8] which says late 1966 for world-wide fall-out. Figure 2 shows the curve obtained from equation (7) compared with the ^{90}Sr mean levels actually measured in soil. The fit to the curve appears to be fairly good, and thus there seems no reason to believe, for the period being, that ^{90}Sr has to any significant degree been removed from Danish soils by processes other than the radioactive decay. The Danish soil mainly consists of clayish loam with a high ion exchange capacity, which undoubtedly reduces the elution of ^{90}Sr (and ^{137}Cs).

Table 1 shows the fall-out rate and the accumulated fall-out in Denmark and the Faroes since 1950. The fall-out rates in Denmark for the years 1950–1954 were estimated from information on nuclear test explosions during those years.[9] For the years 1955–1959 the fall-out rates were estimated from the U.K. data for Milford Haven[10] on the assumption that the ratio between deposited ^{90}Sr activity in

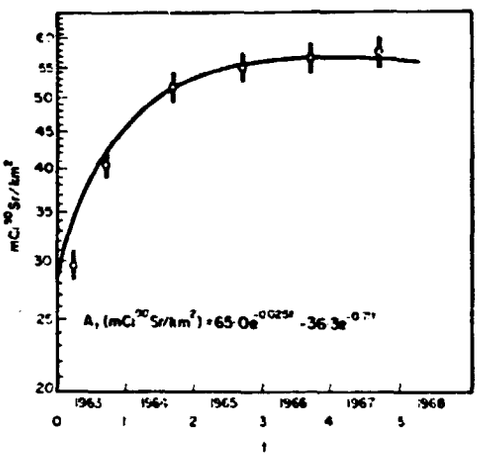

FIG. 2. Accumulated ^{90}Sr in Danish soil (1 SE indicated) compared with the calculated curve (7).

Denmark and at Milford Haven during those years was equal to the mean ratio found in 1962–1967, i.e. 0.82. Since 1960 the Danish fall-out rates are based on values measured during the first years only at one location, Risö, but since 1962 at the ten locations distributed all over the country. The accumulated fall-out was determined from soil measurements carried out in the years 1960–1968.[4] Before 1960 the levels were calculated from the fall-out rates.

Until 1962 the Faroese fall-out rates were estimated from the Danish figures in the table, and so was the accumulated fall-out. Since 1962 the fall-out rate in the Faroes was determined as the mean for the measurements at two Faroese locations.[4] The accumulated fall-out in 1962-1968 was determined from the fall-out rates measured and from the Danish soil data. The fall-out rate in the Faroes in 1962–1968 was 2.2 times the Danish fall-out rate. This ratio has been used for the whole period (1950–1968) during which the Faroese fall-out levels were estimated from the Danish.

Milk

In periods with fall-out rates that are high compared with the accumulated fall-out, the ^{90}Sr levels in Danish milk have been related to

Table 1. ^{90}Sr fall-out levels in Denmark and the Faroes, 1950–1968

	Denmark		The Faroes	
(Yr)	Fall-out rate: (mCi $^{90}Sr/km^2/yr$)	Accumulated fall-out: (mCi $^{90}Sr/km^2$ by the end of the year)	Fall-out rate: (mCi $^{90}Sr/km^2/yr$)	Accumulated fall-out: (mCi $^{90}Sr/km^2$ by the end of the year)
1950	~0.02	~0.1	~0.05	~0.2
1951	~0.1	~0.2	~0.2	~0.5
1952	~0.2	~0.5	~0.5	~1
1953	~0.5	~1	~1	~2
1954	~1	~2	~2	~4
1955	~2.0	~4	~4	~9
1956	~2.0	~6	~5	~13
1957	~2.1	~8	~5	~18
1958	~4.4	~13	~10	~29
1959	~4.7	~18	~10	~40
1960	(1.14)	(19.3)	~2.5	~42
1961	(1.48)	(21.3)	~3.3	~47
1962	7.44	28.7	16.3	62
1963	16.70	44.8	32.7	92
1964	10.41	53.6	25.8	115
1965	3.95	55.3	8.3	120
1966	2.14	56.9	5.4	122
1967	1.02	57.5	3.7	123
1968	1.41	54.4	1.4	121

The figures until 1960 are estimated values. Figures in brackets rely on incomplete data (cf. the text).

the ^{90}Sr levels in air during the year when the milk was produced and the preceding year. A relationship was observed in the years 1959–1964).[11] When the ratio between accumulated fall-out and fall-out rate increases, the indirect contamination of the crops through the roots becomes increasingly important, and the soil factor has to be introduced in the equation along with the rate and lag factors.

Table 2 shows the equations calculated for ^{90}Sr and ^{137}Cs in Danish and Faroese milk for the years 1962–1968. As about half the cows' fodder is produced in the preceding year in Denmark and the Faroes, the fall-out rates both of the year when the milk was produced and the whole of the previous year were taken into account. Although the half-life of ^{137}Cs is a little longer than that of ^{90}Sr, and the $^{137}Cs/^{90}Sr$ ratio in fall-out has varied a little (1.6 ± 0.1) through the years, it was not found worth while to exchange the ^{90}Sr fall-out levels for ^{137}Cs fall-out levels in equations (11)–(13) and (15). In fact the equations show that the ^{137}Cs levels in milk were adequately described by the ^{90}Sr fall-out levels.

It is evident from Table 2 that it was not essential whether concentrations in air or precipitation or deposited activity were used in the rate and lag terms. Concentration instead of deposition data could also be used in these terms for most of the other components of the human food chain. Normally the precipitation data gave a better fit than the air data, probably because precipitation data were based on ten observations, while air data relied on sampling at only one station, Risö. As deposition data are those most widely used in the literature, they will be the only data shown in the remaining equations.

The soil factor in the ^{90}Sr equations for Danish milk was 0.13 pCi $^{90}Sr/g$ Ca per mCi $^{90}Sr/km^2$. The factor was a little higher than the most recent estimate of 0.1 for the U.K.[12] The Faroese soil factor for ^{90}Sr was more than twice the Danish, and this was also the case with the Faroese rate and lag factors. However, the

Table 2. Equations for the prediction of country-wide annual mean levels of ^{90}Sr and ^{137}Cs in Danish and Faroese milk

Equation no.	Area	Equation	Multiple correlation coefficient r	Significance level of r
(8)	Denmark	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.56a_{(i)} + 0.23a_{(i-1)} + 0.14A_{bq(i-1)}$	0.9937	iii
(9)	Denmark	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.65p_{(i)} + 0.30p_{(i-1)} + 0.13A_{bq(i-1)}$	0.9958	iii
(10)	Denmark	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.92d_{(i)} + 0.61d_{(i-1)} + 0.13A_{bq(i-1)}$	0.9965	iii
(11)	Denmark	$(\text{pCi } ^{137}\text{Cs/g K})_{(i)} = 2.18a_{(i)} + 0.51a_{(i-1)} + 0.12A_{bq(i-1)}$	0.9978	iii
(12)	Denmark	$(\text{pCi } ^{137}\text{Cs/g K})_{(i)} = 2.59p_{(i)} + 0.65_{(i-1)} + 0.077A_{bq(i-1)}$	0.9999	iii
(13)	Denmark	$(\text{pCi } ^{137}\text{Cs/g K})_{(i)} = 3.65d_{(i)} + 1.59d_{(i-1)} + 0.057A_{bq(i-1)}$	0.9994	iii
(14)	The Faroes	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 2.43d_{(i)} + 2.09d_{(i-1)} + 0.30A_{bq(i-1)}$	0.9910	iii
(15)	The Faroes	$(\text{pCi } ^{137}\text{Cs/g K})_{(i)} = 10.6d_{(i)} + 11.5d_{(i-1)} + 2.08A_{bq(i-1)}$	0.9827	iii

The milk samples were collected as monthly samples in 1962–1968 from seven dried-milk factories covering the whole country. The Faroese milk was collected weekly as fresh milk from three locations in the Faroes.
a: is the ^{90}Sr level in ground-level air at Risö, in $pCi/10^3m^3$.
p: is the ^{90}Sr mean level in precipitation collected from ten stations all over the country, in pCi/l.
d: is the ^{90}Sr mean deposition from precipitation (from Table 1).
(i): stands for the year (i): $(i-1)$: the preceding year, etc.
A: is the accumulated ^{90}Sr mean level in soil (from Table 1).
1 le milk contains 1.2 g Ca and 1.6 g K.

Table 3. A comparison between observed and calculated milk levels, 1959–68

	Danish milk				Faroese milk			
	(pCi ^{90}Sr/g Ca)		(pCi ^{137}Cs/g K)		(pCi ^{90}Sr/g Ca)		(pCi ^{137}Cs/g K)	
Yr	Observed	Calculated from (10)	Observed	Calculated from (13)	Observed	Calculated from (14)	Observed	Calculated from (15)
1959	(~8.6)	**(8.7)**	(~20)	**(24.9)**	—	**(54)**	—	**(275)**
1960	(4.8)	**(6.2)**	(12)	**(12.7)**	—	**(39)**	—	**(223)**
1961	(4.1)	**(4.6)**	(10)	**(8.3)**	(~28)	**(26)**	—	**(149)**
1962	10.1	**10.4**	31.0	**30.7**	68	**61**	382	**308**
1963	23.8	**23.5**	74.0	**74.5**	131	**132**	627	**663**
1964	24.7	**25.3**	68.0	**67.1**	154	**159**	829	**839**
1965	17.4	**16.7**	33.0	**34.0**	115	**109**	651	**623**
1966	12.0	**11.3**	16.4	**17.2**	73	**67**	417	**402**
1967	9.0	**9.4**	10.1	**10.4**	51	**57**	344	**355**
1968	8.6	**9.2**	11.4	**10.0**	45	**49**	280	**313**

Brackets indicate that the figures are based on incomplete material.

discrepancy between Faroese and Danish agricultural conditions became even more pronounced when ^{137}Cs in milk was considered. The soil factor in Faroese milk for ^{137}Cs was approximately 35 times as high as the soil factor for Danish milk. The estimates of soil factors in the U.K.[12] for ^{137}Cs range from 0.03 to 0.14; the Danish estimate in equation (13) was 0.06. The rate and lag factors in the ^{137}Cs equations also showed a considerable difference between Faroese and Danish conditions (cf. (13) and (15)). Especially the rate factor was considerably higher for the Faroese milk. High soil and lag factors, as we see them in the Faroes, are found in areas with extensive farming and a high organic-matter (and a low mineral) content of the soil.

Prior to 1962 neither fall-out—nor milk data for Denmark were complete, and the data from the period 1959–1961 were thus not included in the calculation of the prediction models. Table 3 shows a comparison between calculated and

Table 4. Exponential equations for the prediction of ^{90}Sr and ^{137}Cs levels in Danish and Faroese milk, 1964–1967

Equation no.	Area	Equation	Reference date	r: multiple correlation coefficient	Significance level of r	Calculated from equations	Remarks
(16)	Denmark	(pCi ^{90}Sr/g Ca) $= 48e^{-0.71t} + 8.3e^{-0.025t}$	1 July	0.9937	ii	5, 7, 10	*The decay of the Faroese ^{90}Sr fallout followed the Danish but the levels were 2.2 times as high
(17)	Denmark	(pCi ^{137}Cs/g K) $= 172e^{-0.71t} + 3.6e^{-0.025t}$	1 July	0.9993	iii	5, 7, 13	
(18)	The Faroes	(pCi ^{90}Sr/g Ca) $= 337e^{-0.71t} + 44e^{-0.025t}$	1 July	0.9801	i	5*, 7*, 14	
(19)	The Faroes	(pCi ^{137}Cs/g K) $= 1650e^{-0.71t} + 301e^{-0.025t}$	1 July	0.9764	i	5*, 7*, 15	

$t = 0$ on 1 January 1963; t is the time in yr. The reference date is the date to which the annual mean is referred, e.g. to obtain the mean level in 1964 t is given the value 1.5, and for 1965 $t = 2.5$ etc. The exponents in the soil factors for Faroese milk are presumably overestimated, as the removal rate of activity from the Faroese soil seems to be greater than for Danish soil [cf. Risö Report No. 181].[16]

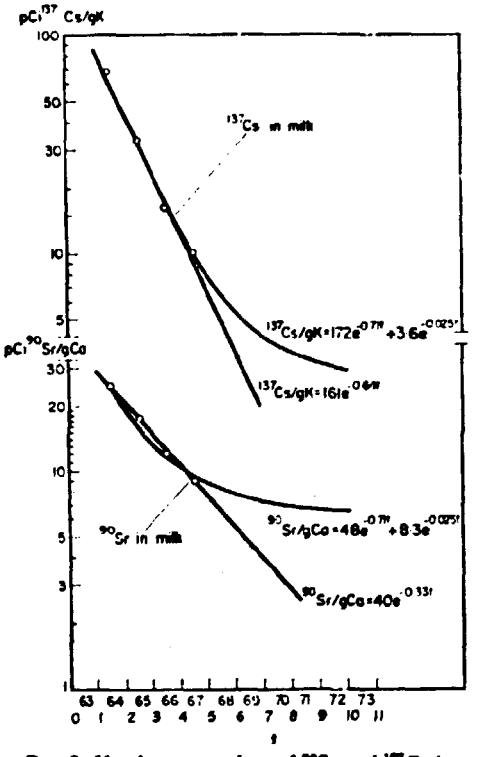

FIG. 3. Yearly mean values of ^{90}Sr and ^{137}Cs in Danish milk compared with the calculated exponential curves (cf. Table 4).

observed milk levels for the period 1959–1968. It is evident that the agreement between observed and calculated values is more satisfactory for the period 1962–1968 than for the first 3 years, but even in 1959–1961 the agreement between observed and calculated values was fairly good.

Table 4 shows the exponential equations calculated for milk, and Fig. 3 shows these double exponential equations compared with single exponential equations calculated from a regression analysis of the Danish milk data from October 1963 to March 1968. During this 4/yr period both ^{90}Sr and ^{137}Cs followed a single exponential decay with effective half-lives of 2.1 and 1.1 yr respectively. If no atmospheric testing had taken place, a deviation from the single exponential decay would, however, have been evident already in 1968 for ^{90}Sr and in 1969 or 1970 for ^{137}Cs owing to the soil uptake.

From Table 4 it can be calculated that in the case of no atmospheric testing since 1962 the ^{137}Cs/g K/^{90}Sr/g Ca ratio would have decreased in Danish milk from 3 to 0.5, while the ratio in the Faroese milk would have increased from 5 to 7; this again demonstrates the completely different behaviour of the activity levels in milk from the two territories.

Cereals

It has been shown[13] earlier that the important period for the contamination of Danish

Table 5. Equations for the prediction of yearly country-wide mean levels of ^{90}Sr and ^{137}Cs in Danish cereals

Equation no.	Species	Equation	Multiple correlation coefficient r	Significance level of r
(20)	Rye	$(pCi\ ^{90}Sr/g\ Ca) = 192d_{(s-a)} + 0.91A_{by(i-1)}$	0.9974	iii
(21)	Barley	$(pCi\ ^{90}Sr/g\ Ca) = 134d_{(s-a)} + 1.05A_{by(i-1)}$	0.9971	iii
(22)	Wheat	$(pCi\ ^{90}Sr/g\ Ca) = 157d_{(s-a)} + 1.30A_{by(i-1)}$	0.9891	iii
(23)	Oats	$(pCi\ ^{90}Sr/g\ Ca) = 67d_{(s-a)} + 0.72A_{by(i-1)}$	0.9880	iii
(24)	Rye	$(pCi\ ^{137}Cs/kg) = 126d_{(m-a)}$	0.9975	iii
(25)	Barley	$(pCi\ ^{137}Cs/kg) = 97d_{(m-a)}$	0.9903	iii
(26)	Wheat	$(pCi\ ^{137}Cs/kg) = 86d_{(m-a)}$	0.9854	iii
(27)	Oats	$(pCi\ ^{137}Cs/kg) = 80.5d_{(m-a)}$	0.9943	iii

The grain samples were collected at the ten locations where precipitation and soil were collected during the the years 1962–1968.
$d_{(s-a)}$: is the mean deposition of ^{90}Sr at the ten locations in July–August.
$d_{(m-a)}$: is the mean deposition of ^{90}Sr at the ten locations in May–August, cf. also the note to Table 1.
The ratio: g Ca/kg grain was for rye: 0.41, barley: 0.47, wheat: 0.40, and oats: 0.82
The ratio: g K/kg grain was for rye: 4.3, barley: 4.6, wheat: 3.8, and oats: 3.8.

cereals with ^{90}Sr is the last months before harvest, i.e. July—August. However, the ^{90}Sr levels in grain also depend on the root uptake of ^{90}Sr from the soil, and equation model (1) was found best suited as prediction model. Table 5 shows the equations for the four grain species. Rye showed the highest rate factor and wheat the highest soil factor. This means that if the fall-out rates are high compared with the cumulative deposit, as was the case in 1962–1964, rye will show the highest levels, but with the increasing importance of soil uptake wheat will show higher levels than rye. The relative ratios between the soil factors in *Table* 5 (0.70: 0.81:1.00:0.55) were nearly identical with the ratios between the relative levels of stable strontium/g of calcium in the four species: (0.67:0.81:1.00:0.52); the multiple correlation coefficient, $r = 0.9990$ was highly significant.

The contamination of Danish grain with ^{137}Cs takes place in May–August,[4] and until now the soil uptake has been negligible. The ^{137}Cs levels in grain were consequently related to the fall-out rate of ^{90}Sr in May–August only. Table 5 shows that, also with respect to ^{137}Cs, rye collects more fall-out by direct contamination than the other species.

The years 1959–1961, which were not included in the material, were tested on the prediction models. Allowing for the fact that the fall-out data for those years were based on observations at one location, the calculated values were in reasonable agreement with the observed levels (cf. Table 6). The mean ratio between calculated and observed values in 1959–1961 was 1.03 (SE:0.06).

Exponential equations could be calculated for cereals for the period 1963–1967 by analogy with the milk equations in Table 4.

Regression analysis of the ^{137}Cs levels in Danish white bread (70% extraction) and rye bread (100% extraction) showed that in the period June 1964–December 1967 the ^{137}Cs levels in these products followed a single exponential decay with an effective half-life of 1.1 yr. In this period the ^{90}Sr in rye bread showed the same half-life. In white bread, however, the ^{90}Sr decayed with a half-life of 1.5 yr, an indication of the importance of soil uptake of ^{90}Sr in the case of wheat.

Vegetables and meat

Vegetables are third in importance as ^{90}Sr donors in the total diet, and meat is nearly as important as milk and cereals as ^{137}Cs donor. Table 7 shows the prediction models for ^{90}Sr in vegetables and ^{137}Cs in meat.

As potatoes show rather high ^{137}Cs levels when fall-out rates are high, prediction models were also calculated for this vegetable. As it was not possible to see any influence of soil uptake on the ^{137}Cs levels in potatoes in the years 1963–1968,

Table 6. Comparison between observed and calculated grain levels, 1959–1968

Year	Rye pCi ^{90}Sr/g Ca Observed	Rye pCi ^{90}Sr/g Ca Calculated from (20)	Barley pCi ^{90}Sr/g Ca Observed	Barley pCi ^{90}Sr/g Ca Calculated from (21)	Wheat pCi ^{90}Sr/g Ca Observed	Wheat pCi ^{90}Sr/g Ca Calculated from (22)	Oats pCi ^{90}Sr/g Ca Observed	Oats pCi ^{90}Sr/g Ca Calculated from (23)
1959	142	(142)*	80	(105)*	100	(124)*	46	(55)*
1960	105	(76)	70	(61)	108	(72)	51	(44)
1961	90	(107)	87	(83)	78	(99)	44	(45)
1962	382	376	296	271	376	320	175	141
1963	1122	1106	766	783	936	922	400	400
1964	485	533	415	391	386	462	177	205
1965	228	212	167	170	215	203	102	96
1966	164	131	123	114	179	138	80	68
1967	73	78	78	79	94	96	52	50
1968	120	135	94	118	126	142	60	70

	Rye pCi ^{137}Cs/kg Observed	Rye pCi ^{137}Cs/kg Calculated from (24)	Barley pCi ^{137}Cs/kg Observed	Barley pCi ^{137}Cs/kg Calculated from (25)	Wheat pCi ^{137}Cs/kg Observed	Wheat pCi ^{137}Cs/kg Calculated from (26)	Oats pCi ^{137}Cs/kg Observed	Oats pCi ^{137}Cs/kg Calculated from (27)
1959	—	(315)	—	(242)	—	(215)	—	(201)
1960	—	(59)	—	(46)	—	(40)	—	(38)
1961	—	(83)	—	(64)	—	(57)	—	(53)
1962	522	533	416	410	363	364	334	341
1963	1220	1256	1038	967	934	857	849	803
1964	840	786	496	605	422	537	439	503
1965	295	255	180	196	166	174	147	163
1966	120	132	73	102	90	90	86	85
1967	56	47	27	36	27	32	29	30
1968	96	107	60	82	53	73	61	68

* The ^{90}Sr deposition in July–August 1959 (0.68 mCi $^{90}Sr/km^2$) was calculated from the ^{90}Sr concentration in air[4] measured at Risø and the amount of precipitation[14] at the ten locations on the assumption that the specific activity in precipitation in pCi/l. was 90% of the air concentration in $pCi/10^3 m^3$ as in 1963–1967 (cf. Fig. 1). In July–August 1960 (0.31 mCi $^{90}Sr/km^2$) and 1961 (0.47 mCi $^{90}Sr/km^2$) the depositions were calculated from the specific activities of precipitation collected at Risø and at the ten locations. The depositions in May–August were calculated in analogy with the estimates for July–August.

potatoes were, as regards ^{137}Cs, related only to the fall-out rate. The soil factors (0.48 and 0.87 on a pCi ^{90}Sr/g Ca basis) for cabbage and carrots (the two most important vegetables in Denmark) were nearly proportional to the stable strontium to calcium ratios (2.5 and 5.1 mg Sr/g Ca) by analogy with the observations for the grain species. The contribution of direct contamination to the ^{90}Sr levels in cabbage and carrots was of minor importance compared with the root uptake.

As the ^{137}Cs levels in meat follow the milk levels,[14] the equation models for milk were also used for meat (cf. Table 7). The lag factor was considerably greater for pork than for beef, in agreement with the fact that pigs eat more stored fodder than cattle. The soil factor for beef was four times as high as that for pigs, which means that in the long run beef will show higher levels than pork when soil uptake is the only source of contamination of the crops.

Table 8 shows that even for the years which

Table 7. Equations for the prediction of the yearly country-wide mean levels of ^{90}Sr and ^{137}Cs in Danish potatoes, cabbage, carrots, and meat

Equation no.	Species	Equation	Multiple correlation coefficient r	Significance level of r
(28)	Potato	$(pCi\ ^{90}Sr/kg)_{(i)} = 0.17d_{(i)} + 0.059A_{bp(i-1)}$	0.8369	i
(29)	Potato	$(pCi\ ^{137}Cs/kg)_{(i)} = 5.45d_{(i)}$	0.9767	iii
(30)	Cabbage	$(pCi\ ^{90}Sr/kg)_{(i)} = 0.44d_{(i)} + 0.24A_{bp(i-1)}$	0.9617	iii
(31)	Carrot	$(pCi\ ^{90}Sr/kg)_{(i)} = 0.58d_{(i)} + 0.27A_{bp(i-1)}$	0.7920	i
(32)	Beef	$(pCi\ ^{137}Cs/kg)_{(i)} = 34.9d_{(i)} + 2.8d_{(i-1)} + 0.64A_{bp(i-1)}$	0.9913	iii
(33)	Pork	$(pCi\ ^{137}Cs/kg)_{(i)} = 34.8d_{(i)} + 22.4d_{(i-1)} + 0.15A_{bp(i-1)}$	0.9866	iii

Potatoes were collected at the ten state experimental farms in 1962–1968 (^{137}Cs was determined only in 1963–1968).

The equations for cabbage and carrots were based on country-wide samplings in 1961–1968 (1962 was missing). Meat was collected in Copenhagen in 1963–1968. As the meat supply of Copenhagen comes from all parts of the country, the Copenhagen meat figures are considered representative of the entire country.

1 kg potatoes contains 0.059 g Ca and 4.0 g K.

1 kg white cabbage contains 0.50 g Ca, and 1 kg carrots contains 0.31 g Ca.

1 kg beef contains 3.6 g K, and 1 kg pork contains 3.6 g K.

Cf. also remarks to Table 1.

were not included when the prediction models were calculated there was a fairly good accordance between observed and calculated values.

Total diet and humans

As the ^{90}Sr levels in total diet follow to some degree the ^{90}Sr levels in milk,[11] the equation model for milk was also applied to total diet (Table 9). The soil factor in the diet equation (34) was close to that for milk (10), so that if all contamination by ^{90}Sr in the human food chain comes from the soil, the pCi ^{90}Sr/g Ca ratio in total diet will nearly equal the level in milk. This observation is in agreement with the estimates made for the U.K.[12]

The best fit to the ^{137}Cs levels in total diet was obtained from equation (35), where the "soil" term was exchanged for the fall-out rate in the year (i-2). If the soil factor was introduced (36), the lag term became the fall-out rate in the year (i-2), while the rate term became the mean of the fall-out rate in the year (i) and the previous year.

Figure 4 shows the relative contribution of ^{90}Sr and ^{137}Cs from the various diet components to the total diet through the years. When the fall-out rate is high as compared with the accumulated fall-out, grain products play a dominating role for ^{90}Sr as well as for ^{137}Cs in Danish diet. When the soil uptake increases, the relative contribution from cereals decreases, while the relative importance of milk products and vegetables (for ^{90}Sr) and of milk products and meat (for ^{137}Cs) increases. As until now no soil factor for ^{137}Cs in cereals has been provable, it is not surprising that equation (36) did not necessarily fit the data for the years 1963–1968 better than (35).

Equations (34) and (35) show that the lag terms were more important than the rate terms; this is in agreement with the fact that a considerable part of the diet (e.g. cereals) consumed in a given year was produced in the preceding year. It is especially the lag factor which causes the difference between the equations for ^{90}Sr in Danish milk (10) and in diet (34).

The ^{90}Sr level in bone from new-borns primarily depends on the ^{90}Sr level in the diet of the mother during the pregnancy. Equation (37) gave the best fit to the data. It should be noted that the lag factor is the fall-out rate in the year (i-2) because the delay is even more pronounced for bone than for diet. From equations (34) and (37) it is possible to estimate the observed ratio (OR) between bone of new-borns and human diet at equilibrium, i.e. at a

Table 8. Comparison between observed and calculated levels in vegetables and meat, 1959–1968

Year	Potato pCi ^{90}Sr/kg Observed	Potato pCi ^{90}Sr/kg Calculated from (28)	Potato pCi ^{137}Cs/kg Observed	Potato pCi ^{137}Cs/kg Calculated from (29)	Cabbage pCi ^{90}Sr/kg Observed	Cabbage pCi ^{90}Sr/kg Calculated from (30)
1959	1.4	(1.6)	—	(26)	—	(5.2)
1960	1.8	(1.3)	—	(6)	(4.0)	(4.8)
1961	(2.0)	(1.4)	—	(8)	4.6	(5.2)
1962	2.5	2.5	—	(55)	(7.6)	8.4
1963	4.3	4.5	100	91	13.6	14.0
1964	4.7	4.4	42	57	15.3	15.1
1965	3.7	3.8	22	22	16.6	14.3
1966	4.3	3.6	11	12	13.0	13.9
1967	3.6	3.5	7	6	13.0	13.8
1968	2.9	3.6	8	8	14.0	14.1

Year	Carrot pCi ^{90}Sr/kg Observed	Carrot pCi ^{90}Sr/kg Calculated from (31)	Beef pCi ^{137}Cs/kg Observed	Beef pCi ^{137}Cs/kg Calculated from (32)	Pork pCi ^{137}Cs/kg Observed	Pork pCi ^{137}Cs/kg Calculated from (33)
1959	--	(6.2)	—	(185)	—	(264)
1960	(5.4)	(5.5)	—	(64)	—	(148)
1961	5.0	(6.1)	—	(67)	—	(80)
1962	(12.3)	10.0	—	(277)	—	(295)
1963	15.6	17.4	630	621	770	752
1964	19.4	18.1	410	439	690	743
1965	22.9	16.7	250	201	470	379
1966	18.0	16.2	110	121	130	171
1967	14.0	15.9	50	78	80	92
1968	11.0	16.3	100	89	80	81

* Brackets indicate that the figures are based on incomplete material.

time when the indirect contamination through the root system is the only source of contamination, $OR_{bone\text{-}diet} = 0.021/0.124 = 0.17$. This ratio should, however, be corrected as the newborns in the material were all from Jutland, where the ^{90}Sr diet levels are approx. 15% higher than in the country as a whole.[4] Hence the OR become 0.17:1.15 = 0.15, which is a little higher than the value observed in the years 1963–1968,[4] perhaps because the foetus also gets some of its bone calcium from the maternal skeleton, which may not have been in equilibrium with the diet.

It was also possible to describe the ^{90}Sr levels in adult vertebrae by an equation similar to that used for newborns. From equations (38) and (34) one can estimate, on the basis of the soil factors, the observed ratio between adult bone and diet at the steady state: OR = 0.032/0.124 = 0.26, which is in agreement with the generally accepted value of 0.25.[15] The Danish diet differs from other western diets for one thing because one third of the diet calcium comes from *Creta praeparata*, and for another because a considerable part of the ^{90}Sr has come from direct contamination of cereals (in 1964 e.g. more than one half). However, in 1961–1967 the ratio between the average integrated diet levels (pCi ^{90}Sr/g Ca) in New York and Denmark was 1.04, while the corresponding ratio for adult vertebrae was 0.98,[16] i.e. the levels from these locations were in close agreement, although the New York diet neither contained *Creta praeparata* nor to any appreciable

Table 9. Equations for the prediction of the yearly country-wide mean levels of ^{90}Sr and ^{137}Cs in total diet and humans in Denmark

Equation no.	Sample	Equation	Multiple correlation coefficient r	Significance level of r
(34)	Diet	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.82d_{(i)} + 1.41d_{(i-1)} + 0.124A_{bot(i-1)}$	0.9926	iii
(35)	Diet	$(\text{pCi } ^{137}\text{Cs/day})_{(i)} = 7.8d_{(i)} + 11.6d_{(i-1)} + 5.5d_{(i-2)}$	0.9988	iii
(36)	Diet	$(\text{pCi } ^{137}\text{Cs/day})_{(i)} = 19.7d_{+(i-1)/2} + 5.8d_{(i-2)} + 0.036A_{bot(i-1)}$	0.9945	iii
(37)	New-born bone	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.168d_{+(i-1)/2} + 0.031d_{(i-2)} + 0.021A_{bot(i-1)}$	0.9799	iii
(38)	Adult vertebrae	$(\text{pCi } ^{90}\text{Sr/g Ca})_{(i)} = 0.0245d_{+(i-1)/2} + 0.059d_{(i-2)} + 0.032A_{bot(i-1)}$	0.9818	iii
(39)	Adult total body	$(\text{pCi } ^{137}\text{Cs/g K})_{(i)} = 6.8d_{+(i-1)/2} + 5.8d_{i-2} + 0.48A_{bot(i-1)}$	0.9973	iii

Total-diet samples were collected in June and Dec. 1962–1968 from 48 towns distributed all over the country. ^{90}Sr has been determined during the whole period, ^{137}Cs since 1963. The yearly diet mean was calculated as: $\frac{\text{diet Dec}_{(i-1)}}{4} + \frac{\text{diet June}_{(i)}}{2} + \frac{\text{diet Dec}_{(i)}}{4}$.

$\frac{i + (i-1)}{2}$: the mean of the current year (i) and the preceding year (cf. also Table 1). The daily intake of calcium is 1.7 g (0.6 g *Creta praeparata*) and the daily intake of potassium 3.8 g. The stable Sr to Ca ratio in the diet was 1.8 mg Sr/g Ca. New-born bones were collected in 1963–1968 only in Jutland from children younger than 30 days. Adult bones were collected all over the country in 1962–1968 from individuals more than nineteen years old. The whole-body measurements have been performed three times a year since 1963 on a control group of approx. sixteen persons at Risö.

degree obtained its ^{90}Sr from direct contamination of cereals. One might argue that the reducing effect on the ^{90}Sr uptake in the bone of the artificially added calcium was less than that of the "naturally" occurring calcium. This would reduce the Danish $OR_{bone/diet}$ value. One might, however, as also suggested by KNIZHNIKOV and MAREI,[17] imagine that ^{90}Sr deposited on the surface of growing grain was absorbed less effectively by the body than ^{90}Sr originating from soil uptake. This would increase the Danish $OR_{bone/diet}$ value. If both these suggestions were correct, the net effect might be that they outbalanced each other. However, if the soil factors in equations (34) and (38) do represent the soil uptake at the steady state, and if the correct $OR_{bone/diet} = 0.25$, it is evident that *Creta praeparata* has the same effect on the ^{90}Sr uptake in the bones as the "natural" calcium in the diet because the availability of ^{90}Sr absorbed through the roots is considered to be the same for all diet components.[18]

The infinite time-integrated level of ^{90}Sr in human vertebrae was calculated, for the years until 1960 from equation (38) and Table 1, for 1961–1968 from the measured values (cf. Table 10) and after 1968 from integration of (43). The integrated level was 2.9 + 14.5 + 73.8 = 91.2 pCi yr/g Ca, neglecting any contribution of ^{90}Sr from explosions since 1962. The dose rate is 0.55 mrad/yr per pCi ^{90}Sr/g Ca to bone marrow and 1.1 mrad/yr per pCi ^{90}Sr/g Ca to endosteal tissue.[16] Hence the dose commitment to Danish bone marrow was: $0.55 \cdot 91.2 = 50$ mrad and that to endosteal cells $1.1 \cdot 91.2 = 100$ mrad. For the northern temperate latitudes UNSCEAR[16] has estimated 64 and 128 mrad respectively. However, this was for an accumulated mean deposited by the end of 1967 of 65

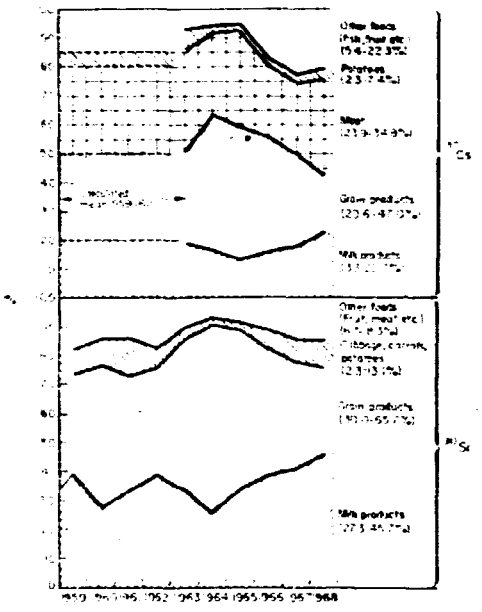

FIG. 4. The relative contributions from the most important ^{90}Sr and ^{137}Cs donors to the activity levels in total Danish diet.

mCi $^{90}Sr/km^2$; as, according to (7), the corresponding Danish accumulated fall-out was only 56 mCi $^{90}Sr/km^2$, the corrected UNSCEAR estimates became 55 and 110 mrads respectively, i.e. in reasonable agreement with the doses calculated above. The total amount of ^{90}Sr injected in the northern hemisphere has been approx. 10 MCi;[16] consequently 1 MCi ^{90}Sr corresponds to a bone marrow dose commitment to the Danish population of approx. 5 mrad. Since the 1961–1962 test series, China has injected approx. 0.6 MCi ^{90}Sr (by 1 October, 1969) in the northern hemisphere. Hence the total dose commitment to the bone marrow becomes 53 mrad fo the Danish population (and 106 mrad to endosteal cells).

UNSCEAR[19] has proposed a formula for the prediction of $^{137}Cs/g$ K in the human body, based on the current yearly fallout and the deposition over the preceding 2 yr. If this formula was used for Denmark, the calculated levels were too high in 1963–1965, while they were too low in 1967–1968.

Equation (39) gave the best fit between observed and calculated data for ^{137}Cs in the total body. As for ^{90}Sr in bone it was most expedient to consider the fall-out rates over a period of three years.

Table 10 shows that the ^{137}Cs levels calculated according to (39) were in excellent agreement with the observed levels. In 1959–1962 there was a fairly good agreement between the calculated UNSCEAR levels and the levels calculated on the basis of (39).

If the contribution from soil uptake to the ^{137}Cs levels in total diet and in the human body were calculated for 1968 according to (35) and

Table 10. Comparison between observed and calculated levels in total diet and humans, 1959–1968

	Total diet				New-born bone		Adult vertebrae		Total body	
	pCi $^{90}Sr/g$ Ca		pCi ^{137}Cs/day		pCi $^{90}Sr/g$ Ca		pCi $^{90}Sr/g$ Ca		pCi $^{137}Cs/g$ K	
		Calculated		Calculated		Calculated		Calculated		Calculated
Year	Observed	from (34)	Observed	from (35)	Observed	from (36)	Observed	from (37)	Observed	from (38)
1959	(9.9)	(11.7)	—	(99)	—	(1.1)	—	(0.7)	—	(49)
1960	(7.9)	(9.8)	—	(88)	(0.7)	(1.0)	(0.6)	(0.9)	—	(54)
1961	5.9	(5.2)	—	(51)	—	(0.5)	0.8	(0.9)	—	(46)
1962	10.7	10.8	—	(81)	—	(1.2)	0.8	0.9	—	(47)
1963	27.5	27.7	223	224	2.8	2.7	1.2	1.3	100	104
1964	38.5	37.6	318	315	3.3	3.4	2.4	2.2	162	157
1965	22.5	24.5	236	244	2.9	2.8	2.7	2.9	170	172
1966	16.5	14.2	128	120	1.9	2.0	2.6	2.5	106	108
1967	10.5	10.9	62	55	1.8	1.6	2.1	2.1	65	61
1968	9.2	8.5	35	35	1.3	1.5	1.9	2.0	46	48

Figures in brackets are based on incomplete material.

Table 11. Exponential equations for the prediction of ^{90}Sr and ^{137}Cs levels in total diet and humans in Denmark

Equation no.	Sample	Equation	Ref. date	Multiple correlation coefficient r	Significance level of r	Calculated from equations	Valid for the period
(40	Total diet	(pCi ^{90}Sr/g Ca) $= 86.8e^{-0.71t} + 8.1e^{-0.025t}$	July 1	0.9973	—	(5), (7), (34)	1964–1967
(41)	Total diet	(pCi ^{137}Cs/day) $= 1370e^{-0.71t}$	July 1	0.9984	—	(5), (35)	1965–1967
(42)	New-born bone	(pCi ^{90}Sr/g Ca) $8.2e^{-0.71t} + 1.37e^{-0.025t}$	July 1	0.9927	—	(5), (7), (37)	1965–1967
(43)	Adult vertebrae	(pCi ^{90}Sr/g Ca) $5.43e^{-0.71t} + 2.11e^{-0.025t}$	July 1	0.8724	—	(5), (7), (38)	1965–1967 1965–1967
(44)	Adult whole body	(pCi ^{137}Cs/g K) $= 830e^{-0.71t} + 31.5e^{-0.025t}$	July 1	0.9987	—	(5), (7), (39)	1965–1967

Cf. notes to Table 4.

(39) it resulted in the soil contribution being 6% for the diet but 58% for the body. The percentage for milk was 33, for beef 45 and for pork 10. It is thus evident that the soil factor for total diet was too low while that for the human body was too high. The cause of this discrepancy might be that the influence of the soil uptake on the ^{137}Cs levels in the human food chain was so small that reliable determinations of the soil factors for ^{137}Cs have until now been impossible. There might, however, be other explanations. Approximately 8% of the ^{137}Cs body burden have been estimated to be present in bone.[16] If the biological half-time of ^{137}Cs is long in bone [18.9 yr][29] one would expect to see the body burden decrease more slowly with time. A slower decrease would result in the estimation of a greater soil factor in equation (39).

A regression analysis of the ^{137}Cs levels in diet for the period December 1963–December 1967 showed that the levels followed a single exponential decay curve with a half-life of 1.25 yr. The total-body levels decayed from 1965 with a half-life of 1.5 yr. These half-lives are in agreement with observations by GUSTAFSON.[21]

The Danish ratio between human body and diet levels (pCi ^{137}Cs/g K figures) was 3:1 (cf. Table 10) i.e. also in accordance with US observations.[29]

By analogy with ^{90}Sr the infinite-time-integrated level of ^{137}Cs in the total body was calculated from (39), Table 10, and (44) as $307 + 649 + 1122 = 2078$ pCi ^{137}Cs yr/g K. According to UNSCEAR[16] the dose rate from 1 pCi ^{137}Cs/g K in the total body of a man weighing 70 kg is 18 μrad/yr. Hence the dose commitment to the total adult body was 37 mrad, this figure is considered an upper estimate as the soil factor in (44) is probably overestimated. As a lower estimate one might use (41) instead of (44) in the calculation of the dose after 1968, on the assumption that the ^{137}Cs/K ratios between human body and diet were 3:1. In that case the dose commitment is 18 mrad. UNSCEAR[16] has estimated 21 mrad in the northern hemisphere; if this figure was corrected according to Danish deposition conditions (cf. above), the estimate would be 18 mrad, i.e. equal to the lower Danish estimate above.

^{137}Cs has been decreasing in all diet components since 1963 except in fish, where the levels have shown a slight increase through the years. That means that the contribution of ^{137}Cs from fish has been increasing since 1963–1964. In 1968, 7% of the total ^{137}Cs intake came from fish. The long term implication of this in the equations for ^{137}Cs in diet will be a more pronounced soil factor than displayed in equation (36). If no fresh fall-out had occurred since 1962 it may be estimated that the contribution from fish to total-diet ^{137}Cs in 1972 would have equalled the contribution from all other diet components.

CONCLUSION

For the most important components in the human food chain and for the total diet it has been possible to describe the ^{90}Sr and ^{137}Cs levels on the basis of fall-out rate and accumulated deposit as independent variables. The fall-out rate could be represented either by deposition data (mCi $^{90}Sr/km^2/yr$) or by precipitation or ground-level air concentrations (pCi $^{90}Sr/l$ or pCi $^{90}Sr/10^3$ m^3). The accumulated deposit was measured as the soil activity down to a depth of 20 cm (mCi $^{90}Sr/km^2$). In most cases it was found expedient to split up the fall-out rate into two terms: the rate for the current year and the rate for the preceding year. The accumulated deposit was always referred to the end of the preceding year. After the 1961–1962 test series the ^{90}Sr levels in air and precipitation followed until 1968 an exponential decay with a half-life of approximately 10 months. This resulted in an almost exponential decay for most diet components during the years 1964–1967 with half-lives from 10 to 25 months depending on the importance of direct and indirect contamination. ^{90}Sr in milk e.g. decayed with a half-life of a little over 2 yr, while ^{137}Cs in rye showed a half-life of somewhat less than 1 yr. If no atmospheric testing had been performed, a deviation from a single exponential decay of diet levels would have appeared in 1968–1969 for ^{90}Sr and at the beginning of the seventies for ^{137}Cs.

The influence of soil uptake has been so marked for ^{90}Sr in the last few years that reliable determinations of the soil factors in the equations have been possible. By means of these soil factors the OR value for adult bone/total diet was estimated at 0.26, and for foetal bone/mothers' diet at 0.15, i.e. both ratios are in reasonable agreement with estimates made by other methods.

The *Creta praeparata* and the considerable amounts of ^{90}Sr from direct contamination of cereals in the Danish diet had no observable effects on the Danish $OR_{diet/bone}$ values.

The soil factors in grain species and vegetables were proportional to the stable strontium to calcium ratios in the respective species.

Although the soil factors for ^{90}Sr seem reliable, little is known of the long-term effect on the availability to crops of ^{90}Sr bound to the soil minerals. Furthermore no reliable estimate of the removal rate of ^{90}Sr by weathering processes exists. At least no such effect has until now been detected under Danish agricultural conditions. It is therefore considered justifiable to continue the surveys of ^{90}Sr in the human food chain.

As regards ^{137}Cs, the significance of the soil factors is more doubtful because the influence of soil uptake of ^{137}Cs has until now been modest. It is thus indicated that the ^{137}Cs investigations should also be continued in the years to come to obtain a more reliable determination of the soil factors for ^{137}Cs and thus of the long-term fate of ^{137}Cs in the human food chain.

The ^{90}Sr as well as the ^{137}Cs human levels have followed equations similar to those calculated for the diet. The equation for whole-body ^{137}Cs showed a surprisingly high soil factor, which is supposed to be due to an accumulation of ^{137}Cs in bone.

While Denmark is an area with intensive farming, high mineral (clay) content of the soil and medium precipitation (650 mm/yr), the Faroes are characterized by extensive farming, low mineral and high organic-matter content of the soil and a high precipitation level (1500 mm/yr). A comparison of the prediction models for milk from the two areas shows marked differences: The soil factors in the Faroes were considerably greater, especially as regards ^{137}Cs and the lag as well as the rate factors also exceeded the Danish values.

From integration of the exponential equations in Table 11 and from UNSCEAR's information on dose-rate factors[17] the integrated average dose to members of the Danish population over an infinite time was calculated. The dose commitment from ^{90}Sr to bone marrow was 53 mrad and to endosteal cells 106 mrad. The whole-body dose commitment from ^{137}Cs was as an upper estimate calculated to be 37 mrad from whole-body measurements and as a lower estimate 18 mrad from total diet measurements.

REFERENCES

1. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. New York (1958).
2. B. O. Bartlett and R. Scott Russell, *Nature Lond.* **209**, 1062 (1966).
3. HASL-210, pp. 1-4 (1969).

4. Risö Reports Nos. 9, 14, 23, 41, 63, 64, 85, 86, 107, 108, 130, 131, 154, 155, 180, 181, 201 and 202 (1959–69).
5. R. G. Osmond, M. J. Owers, C. Healy and A. P. Mead, AERE-R 2899 (1959).
6. F. J. Bryant, A. Morgan and G. S. Spicer, AERE-R-3030 (1959).
7. HASL-204, pp. 1–4 (1969).
8. E. P. Hardy, M. W. Meyer, J. S. Allen and L. T. Alexander, *Nature, Lond.* **219**, 584 (1968).
9. HASL-142, p. 218 (1964).
10. R. S. Cambray, E. M. R. Fisher, W. L. Brooks and D. H. Peirson, AERE-R-5899 (1968).
11. A. Aarkrog, *Acta Radiol.* (*Supplementum*) **254**, 13 (1966).
12. ARCRL 19, p. 12 (1969).
13. A. Aarkrog, *Proc. 1st Intern. Congr. Radiation Protection*, Rome, 1966, pp. 1047–1063 (1968).
14. Bo Lindell and Attilio Magi, *Ark Fys* **29**, 69 (1965).
15. C. L. Comar, *Strontium Metabolism* (Edited by J. M. A. Lenihan, J. F. Loutit and J. H. Martin) pp. 17-31, Academic Press, New York (1967).
16. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, New York (1969).
17. V. A. Knizhnikov and A. N. Marei, *Strontium Metabolism* (Edited by J. M. A. Lenihan, J. F. Loutit and J. H. Martin) pp. 71–82 (1967).
18. T. E. F. Carr, G. E. Harrison, J. F. Loutit and A. Sutton, *Nature, Lond.* **196**, 200 (1962).
19. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, New York (1966).
20. P. F. Gustafson and J. E. Miller, *Health Phys.* **16**, 167 (1969).
21. P. F. Gustafson, J. S. Brar and D. M. Nelson, HASL-204, 1-78 (1969).

Health Physics Pergamon Press 1968. Vol. 15, pp. 105–114. Printed in Northern Ireland

STRONTIUM-90 IN SHED DECIDUOUS TEETH COLLECTED IN DENMARK, THE FAROES AND GREENLAND FROM CHILDREN BORN IN 1950–1958

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Abstract—Shed deciduous teeth were collected in 1962–1965 from 25 schools in Denmark, the Faroes and Greenland. Approx. 10,000 teeth from children born in the period 1950–1958 are included in the present investigation, which compiles 215 ^{90}Sr analyses. The ^{90}Sr level in deciduous teeth crowns increased by a factor of four from 1953 to 1958. Faroese teeth contained nearly 4 times as much ^{90}Sr as teeth from Denmark and Greenland. Incisors contained approx. three fourths of the level found in second molars. The ^{90}Sr diet levels measured in 1962–1966 in Denmark, the Faroes and Greenland showed a similar variation between locations as the ^{90}Sr in deciduous teeth from children born in 1950–1958 in the same areas. The ^{90}Sr mean level in shed deciduous teeth is considered as an estimate of the ^{90}Sr content in the tooth donors bone when he was 1 year old.

INTRODUCTION

As SHOWN by several authors[1–3] the tooth crown provides information on the dietary ^{90}Sr at the time of tooth formation. The mineralisation of deciduous tooth crowns takes place in the last months of the pregnancy and within the first year after birth[4] and the ^{90}Sr levels in shed deciduous teeth should thus be related to the ^{90}Sr diet levels of the mother during the pregnancy and the infant during the first year of life.

The purpose of the present study is to compare the ^{90}Sr levels in deciduous teeth collected in five different areas: three representing Denmark and the remaining two, the Faroes and Greenland.

From the measurements, the intention is also to obtain an estimate of the ^{90}Sr levels in infants' bone especially from the Faroes and Greenland. The population numbers in these areas are rather low (approx. 36,000 in each country), it has therefore been impossible to obtain a sufficient number of bone samples for a reliable evaluation of the ^{90}Sr levels in human bone. The measurements of ^{90}Sr in foodstuffs[5,6] and human teeth are, thus far, the only way of estimating the human bone levels in the Faroes and Greenland

MATERIALS AND METHODS

The material consists of approx. 10,000 shed deciduous teeth obtained in the years 1962–1965 through smaller local schools in rural districts (cf. Fig. 1). The idea was that contact, which was supposed to last several years, was easy to maintain with a small school.

The schools collected the teeth from pupils born in the district where the school was located. To stimulate the interest for the tooth sampling among the children each tooth was paid with 50 Danish øre (approx. 7 U.S. cts). The teachers pooled the teeth in polyethylene bottles marked with the birth years of the tooth donors. Twice a year the samples were sent to Risö, and grouped according to area (Jutland, The Islands, Bornholm, The Faroes and Greenland) birth year (1950, 1951–1958), and tooth type (incisors, cuspids, first molars and second molars). After the sorting, the teeth were ashed at 700°C, then roots and fillings were removed. Incisors and cuspids were normally sound whereas the molars mostly were carrious. Before the radiochemical analysis the ashed teeth were crushed in a mortar and the ash was blended and weighed.

106 ^{90}Sr IN DECIDUOUS TEETH COLLECTED FROM CHILDREN BORN IN 1950–1958

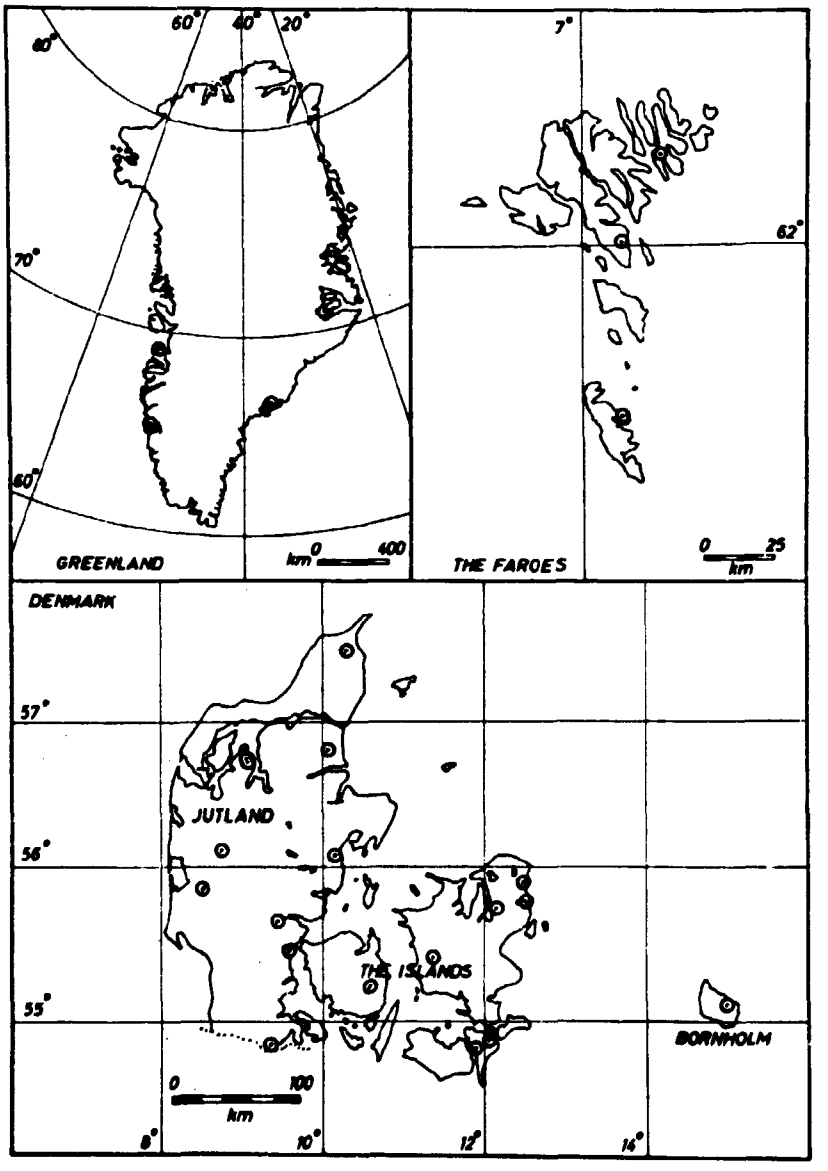

FIG. 1. Sampling locations for teeth.

215 ^{90}Sr-analyses are included in this investigation. The average ash weight for a ^{90}Sr determination was approx. 9 g. Analyses of samples from children born before 1954 normally contained more ash than this average, while samples after 1957 in some cases contained only 2–3 g ash. The tooth analyses from Faroese children contained as a whole less ash than those from Greenland and Denmark, because the Faroese ^{90}Sr levels were expectedly higher.

^{90}Sr was determined according to the classical radiochemical strontium analyses[7] using fuming nitric acid separations and a final milking and counting of the ^{90}Y-daughter. Calcium was determined in a number of pooled samples by precipitation and weighing of the oxalate.[7] The mean content was 0.385 g Ca per gram of tooth ash (S.D. 0.010 g Ca).

Of the analyses 25 % were blanks consisting of 5 g $Ca_3(PO_4)_2$. Most of the samples were measured in automatic sample changers taking 4 samples each, one of the samples being a blank used as background for the other samples. The counters in the sample changers were anti-coincidence Geiger Flow-Counters[6] with a 25 mm ø mylar foil window (0.9 mg/cm^2). The background was approx. 0.3 counts/min and the mean counting efficiency for ^{90}Y was 0.36 counts/min per dis/min.

68 samples which yielded bad decay curves for ^{90}Y, were also counted as (^{90}Sr + ^{90}Y), and only the cases (54 samples) where this counting fitted the ^{90}Y counting were included in the material.

As the samples were counted 10–15 years after the ^{90}Sr was incorporated in the tooth tissue, all samples were corrected for decay of ^{90}Sr ($t_{\frac{1}{2}}$ = 28 yr) back to the birth year of the child. Samples which showed no detectable ^{90}Sr content (indicated by an asterisk in Table 1) were reported as one standard deviation of the determination.

The logarithm of the figures are approximately normally distributed and the results have consequently been transformed to their natural logarithms before the statistical treatment. The analysis of variance, used in this treatment, has been programmed for a computer.[9] The programme is able to calculate missing values in the material, and furthermore gives complete tables, when all interactions equal zero, i.e. a material where only the main effects are reflected (cf. Fig. 2).

RESULTS AND DISCUSSION

Table 1 shows the decay corrected pCi ^{90}Sr/g Ca levels in the samples, and Table 2 the analysis of variance of the natural logarithm of the figures.

Variation with birth year

In accordance with observations of other authors[2,10–12] the ^{90}Sr concentration in the teeth, as shown in Fig. 2 has been continuously increasing throughout the years. It was however, not possible to prove a significant difference between teeth from 1956, 1957 and 1958 (the difference between 1956 and 1958 was significant only at the 95 % level).

The first thermonuclear test was performed on 31 October 1952. Until this date the total fission yield of air explosions carried out during 1945–1951 had been approx. 0.2 MT, of which more than 0.1 MT were detonated in 1951.[13]

If the 0.02 MCi ^{90}Sr[14] created in the period 1945–1951 had been evenly distributed exclusively over the temperate zone in the northern hemisphere, and if half of the ^{90}Sr, i.e. 0.01 MCi, was deposited in 1951, then the accumulated fallout in the northern temperate zone by the end of 1951 would have been approx. 0.2 mCi ^{90}Sr/km^2. The fallout rate would have been approx. 0.1 mCi ^{90}Sr/km^2/yr in 1951 and 0.02 on the average for the period 1946–1950.

If these fallout levels were valid for Denmark and if the specific ^{90}Sr activity found in precipitation at Milford Haven in the U.K.[15] in the period 1955–1958 equals the Danish rain-water concentrations in this period then the fall-out levels in 1955–1958 could be calculated from the U.K. data[15] and from Danish data[16] on amounts of precipitation. The 1952–1954 fallout rates in Denmark were estimated from the information on announced nuclear detonations[13] and from the U.K. information on the amount of accumulated fallout up to the end of 1954.[15]

Table 3 shows the estimated fallout levels based on the above mentioned assumption. The milk levels in the table were calculated from:

$$C_m = 0.77\,F_r + 0.10\,F_d \qquad (1)$$

108 ^{90}Sr IN DECIDUOUS TEETH COLLECTED FROM CHILDREN BORN IN 1950–1958

where C_m is pCi ^{90}Sr/g Ca in milk produced in the year (i)

F_r is the fallout rate: mCi ^{90}Sr/km^2 per 2 years in the years $(i-1)$ and (i)

F_d is the accumulated fallout by the end of year $(i-1)$ and 0.77 and 0.10 are empirical proportionality factors found valid in Denmark for the period 1960–1966.[17]

The ^{90}Sr in shed deciduous teeth is incorporated in the tooth tissue in the last months of the pregnancy and within the first 12 months of

Table 1. pCi ^{90}Sr *in shed deciduous teeth from Denmark, The Faroes and Greenland*

Birth year of the cohort		1950	1951	1952	1953	1954	1955	1956	1957	1958
		0.034*	0.071*	0.048*	0.39	0.94	1.18	0.78	0.87	1.39
	Incisors				0.97	0.82	0.86		1.34	2.06
						0.70	0.89			
		0.094	0.15	0.31	0.32	0.62	0.70	1.61	1.42	1.08
	Cuspids			0.67	0.81	1.22	1.24	1.26	1.40	
					0.70	0.92				
Jutland		0.024*	0.46	0.19	0.38	0.70	0.88	1.30	1.51	
	Molar 1			0.42	0.62	0.79	1.05	0.75	1.88	
				0.64	0.55	1.01				
			0.24	0.40	0.72	0.60	0.85	1.10	2.01	3.66
	Molar 2			0.33	0.61	0.79	1.19	1.57	2.30	
					0.60	1.05	1.18	1.50		
	Incisors					0.55	0.78	0.95	1.26	1.46
										1.77
		0.14	0.14		0.60	0.36	0.90	1.58		
	Cuspids				0.45	0.85	1.06	1.13		
					0.28	0.58				
The Islands			0.19		0.54	0.52	0.74	1.04	1.84	0.91
	Molar 1				0.25	0.80	0.88	1.21	0.57	1.60
					0.26	0.87				
			0.38	0.35	0.80	0.49	1.25	1.10	2.69	1.55
	Molar 2		0.25	0.22		0.52	1.36	2.82		
							0.73	1.26		
	Incisors				0.34	0.49	0.92	0.96	1.03	1.62
								1.39	1.33	
	Cuspids		0.12	0.30	0.34	1.02	1.21			
							1.04			
Bornholm	Molar 1		0.13	0.28	0.40	0.67				
	Molar 2	0.09	0.11	0.63	0.55	1.10	1.04			
					0.41	0.69				
	Incisors	0.22	0.62	0.83	1.67	2.42	3.07	4.01	4.97	
									5.86	
	Cuspids	0.22	0.54	0.98	1.16	1.60	2.73	3.34	5.75	
					2.61	3.77	4.88	5.02		
The Faroes	Molar 1	0.28	0.39	2.64	0.76	1.68	0.58	2.84	4.90	
				4.16	2.82	3.02	3.86	6.05	6.24	
	Molar 2	0.77	0.55	1.19	1.30	2.24	3.89	4.15	5.43	
				1.92	2.06	2.62	4.70	7.11	5.91	
	Incisors					1.06	0.77	1.06	1.11	1.43
Greenland	Cuspids			0.34	0.26	0.70	1.14	1.05	1.73	1.63
	Molar 1			0.30	0.26	0.50	1.14	1.36	1.05	
	Molar 2			0.29	0.32	0.52	0.84	1.24	1.28	

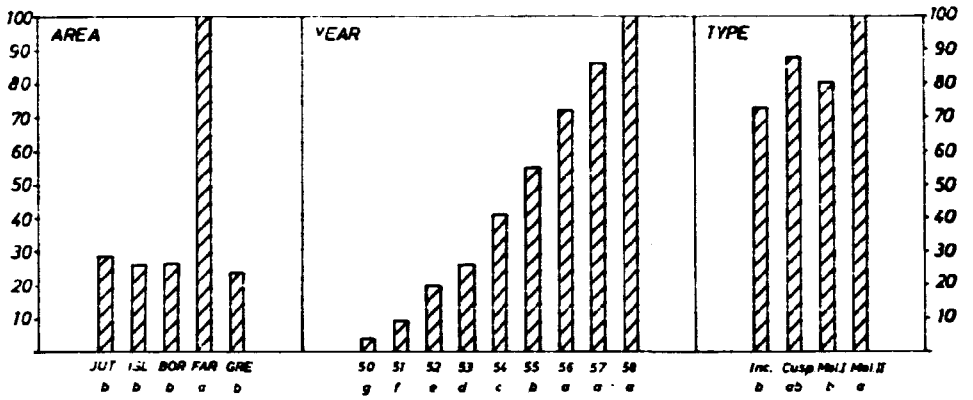

FIG. 2. Relative pCi ^{90}Sr/g Ca levels in shed deciduous teeth, from Denmark, The Faroes and Greenland from children born in 1950–1958. Columns with different small letters are significantly different ($P > 99\%$).

Table 2. Analysis of variance of the ln pCi ^{90}Sr/g Ca *in shed deciduous teeth (cf. Table 1) from children born in* 1950–1958 *in Denmark, The Faroes, and Greenland*

Effect	Source	SSD	f	s^2	v^2	P
Main	Tooth type	2.6440	3	0.8813	5.70	>99.9%
	Area	63.5524	4	15.8881	102.84	>99.95%
	Birth year	124.0016	8	15.5002	100.32	>99.95%
2 factor interaction	Type-Area	1.3490	12	0.1124	0.83	—
	Area-Year	4.8750	29	0.1681	1.24	—
	Year-Type	6.4666	24	0.2694	1.99	>99%
3 factor interaction	Type-Area-Year	6.9609	56	0.1243	0.87	—
Remainder		11.1848	78	0.1434		

The 3 factor interaction was tested against remainder ($f = 78$), the 2 factor interaction against remainder and 3 factor interaction ($f = 134$), and the main effects against remainder and all interactions $f = 199$).

Table 3. Estimated ^{90}Sr *levels in fallout and milk in Denmark* 1950–1958

Year	Estimated annual fallout rate (mCi ^{90}Sr/km^2/yr)	Estimated accumulated fallout F_d (mCi ^{90}Sr/km^2)	Calculated milk levels from fallout data (equation (1)) C_m (pCi ^{90}Sr/g Ca)
1950	0.02	0.08	0.04
1951	0.1	0.1	0.1
1952	0.1	0.2	0.2
1953	0.4	0.3	0.4
1954	0.8	0.7	1.0
1955	1.7	1.5	2.1
1956	1.8	3.1	3.0
1957	2.0	4.8	3.4
1958	3.4	6.6	4.8

life. REISS[1] has estimated the portion of shed tooth formed at birth. She found for incisors: 32%, for cuspids: 6%, for first molars: 17%, and for second molars: 5%. Approx. 15% of the average shed tooth is thus formed prenatally and 85% postnatally. The observed ratio (OR) between fetal bone and mother's diet is 0.09[17] and the bone-diet OR for infants (0–1 yr) is estimated by HASL[18] at 0.35. If it is assumed that the infant's diet during the first year of life corresponds to 75% milk and 25% total (adult) diet, the following equation could be set up:

$$C_i = 0.15 \cdot 0.09\, C_{D_1} + 0.85 \cdot 0.35\, (0.75\, C_M + 0.25\, C_{D_2}) \quad (2)$$

where C_i is pCi ^{90}Sr/g Ca in shed deciduous teeth from children born in the year (i)

C_{D_1} and C_{D_2} are mean pCi ^{90}Sr/g Ca in total diet in the years (i) and ($i - 1$) and in the years (i) and ($i + 1$) respectively

C_M is mean pCi ^{90}Sr/g Ca in cow milk in the years (i) and ($i + 1$)

$C_D = 2\, C_M$ in 1950–57 and $C_D = 1.5\, C_M$ since 1958[17] (due to the addition of creta praeparata ($CaCO_3$) to the Danish flour since April 1958).

Table 4 shows the ^{90}Sr levels in shed deciduous teeth from Denmark along with the calculated levels from equation (2).

The equation underestimates the tooth levels in the first years of the period. REISS,[1] and later ROSENTHAL,[10] have assumed that the average concentration of ^{90}Sr in the teeth of children born in 1951 and 1952, when fallout was low, might represent ^{90}Sr appearing in teeth as a result of direct exchange with dietary ^{90}Sr, during the 5–7 years the teeth remained in the body before being shed. The average concentration in the teeth from 1951 and 1952 was 0.18 pCi ^{90}Sr/g Ca in the St. Louis material; Reiss subtracted this figure from her results as a correction for external accretion of ^{90}Sr after the formation of the teeth. It could, however, be questioned whether such a constant correction is proper, as the mean diet levels in the seven year periods: 1950–1956 and 1958–1964 differed by more than a factor of ten. PETERSEN *et al.*[19] found little or no evidence for an accretion of ^{226}Ra in deciduous teeth after formation. Recently LINDEMANN[20] has reported deciduous tooth crowns extracted in 1964 showing higher ^{90}Sr levels than teeth extracted in 1963, although the teeth were from children born in the same year. He suggested that ^{90}Sr is possibly not so easily resorbed as calcium, or that resorbed ^{90}Sr is re-captured by unresorbed hard tooth tissues. In addition the possibility that unresorbed hard tooth tissues in the teeth undergoing resorption, are susceptible to continued supply of ^{90}Sr, may explain the increased amount of ^{90}Sr per gram calcium in this period.

If a constant correction of 0.2 pCi ^{90}Sr/g Ca for a possible external ^{90}Sr accretion is subtracted from all the mean tooth levels in Table 4, the agreement between measured and calculated tooth levels would be improved in the first part,

Table 4. Measured and calculated pCi ^{90}Sr/g Ca *levels in shed deciduous teeth collected in Denmark*

Birth year (i)	The mean level C_i in shed deciduous teeth and the standard error of the mean (from Table 1)	Number of samples in the mean	C_i calculated from equation (2) (cf. Table 3 and Ref. 17)
1950	0.06 ± 0.03	5	0.03
1951	0.20 ± 0.04	11	0.06
1952	0.36 ± 0.05	13	0.1
1953	0.52 ± 0.04	23	0.3
1954	0.76 ± 0.04	26	0.6
1955	1.00 ± 0.04	22	1.0
1956	1.30 ± 0.11	18	1.3
1957	1.53 ± 0.15	14	1.5
1958	1.71 ± 0.24	10	2.0

but depreciated at the end of the period. The discrepancy in 1957–1958 might be due to the fact that the sample-proportion of second molars and cuspids, (which show the highest levels (cf. Fig. 2)) was lower in these years than in the preceding period.

In conclusion: The present material does not exclude the possibility of an external accretion of ^{90}Sr after tooth formation, but it does not seem to be an important factor for the ^{90}Sr levels in teeth formed since 1954.

The interaction between birth year and tooth type was significant (cf. Table 2). This is explained by the differences in postnatal calcification times for the different tooth types, (the differences in prenatal calcification are less important, due to the higher discrimination against ^{90}Sr in utero). As cuspids and molars depend upon the dietary ^{90}Sr levels after birth for a longer period than incisors,[1,4] changes in these levels might alter the ^{90}Sr concentrations in the former without a corresponding change in the latter. Figure 3 shows this effect: while incisors display increasing activity throughout the period, cuspids and molars show a flattening out by the end of the period.

Variation with tooth type

As shown in Fig. 2 the ^{90}Sr concentration was significantly higher in second molars than in incisors and first molars, and cuspids probably contained ($P > 95\%$) more activity than incisors. This picture fits the aforementioned model proposed by REISS[1] for the calcification of shed deciduous teeth. As the prenatal discrimination against ^{90}Sr is greater than the postnatal, it was to be expected, in a period with increasing ^{90}Sr diet levels, that the ^{90}Sr concentrations in the four types of teeth were related as: Second molar > cuspids > first molar > incisors, which agrees with Fig. 2. ROSENTHAL *et al.*[12] found only minor differences in the ^{90}Sr content of various teeth, although the authors do comment, that it might have been expected, due to the longer development time of cuspids and deciduous second molars which would result in the deposition of substantially more ^{90}Sr than in incisors and deciduous first molars.

Variation with birth location

Deciduous teeth from the Faroes contained nearly four times as much ^{90}Sr as teeth from Denmark and Greenland. In Fig. 4 the relative pCi ^{90}Sr/g Ca levels in total diet and in milk from Denmark, the Faroes and Greenland is shown for the period 1962–1966.[5,6,17] The total diet in the Faroes and Greenland is only partly locally produced; all milk in Greenland and 25% of the Faroese milk is imported from Denmark. If the model for diet composition applied in equation (2) is used for a comparison of the diet levels determining the shed deciduous tooth levels in the five areas considered, and if Fig. 4 is used, the relative levels in such a diet is in Jutland: 29, in the Islands: 19, in Bornholm: 20, in the Faroes: 100, and in Greenland: 23. These values agree reasonably well with the relative tooth levels shown in Fig. 2.

Breast feeding for longer periods after birth is not very common in Denmark, but is somewhat more widespread in Greenland. This[12] along with lower total diet levels might explain the lower mean level in Greenland teeth than in Danish, although the difference is not statistically significant. As indicated in Fig. 2 the difference between the tooth levels in the three areas in Denmark was also not significant. It is however, interesting from Fig 4 to note a similar trend in diet levels as in tooth levels: Jutland > Bornholm > The Islands. From measurements of adult human bone from 1960–1966,[17] it can be shown, that the bone level in Jutland was approx. 20% higher than in the Islands. The difference is mainly due to the higher amounts of precipitation and hence of fallout in Jutland (approx. 20% higher than in the Island.[17] It should, however, also be noticed, that while the soils in Jutland preferentially are sandy, the Islands have soils rich in clay. This results in higher ^{90}Sr uptake in the crops in Jutland as compared with the Islands. Differences in tooth levels between areas with different amounts of precipitation have previously been observed in the U.K.[3]

The great difference between Faroese and Danish tooth levels is only partly due to the fact, that the ^{90}Sr fallout is 2–3 times greater in the Faroes than in Denmark.[5,17] The extensive farming methods applied in the Faroes

112 ^{90}Sr IN DECIDUOUS TEETH COLLECTED FROM CHILDREN BORN IN 1950–1958

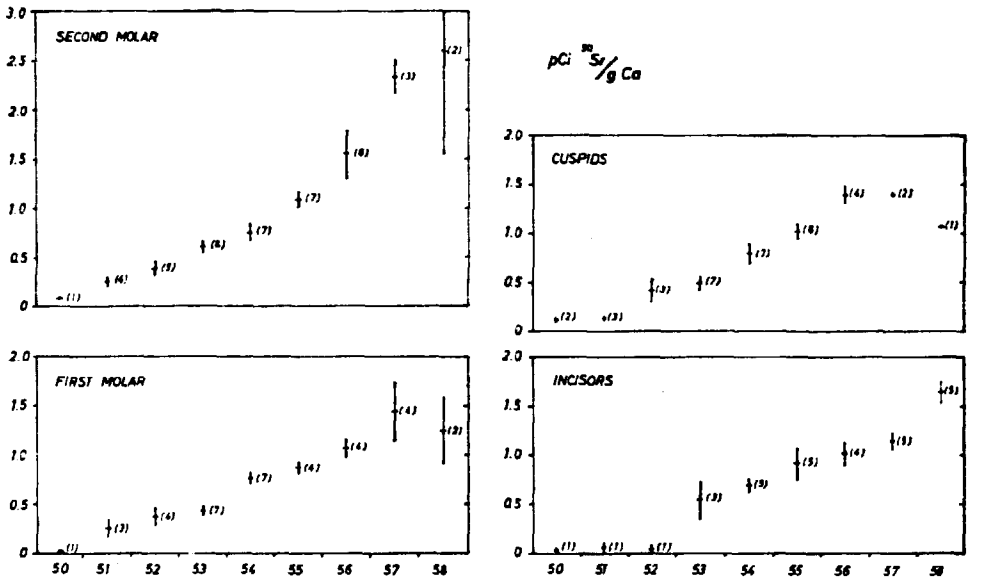

FIG. 3. Mean concentrations of ^{90}Sr in Danish deciduous teeth. The standard errors of the means are indicated in the figures. The numbers of samples included in the mean are shown in brackets.

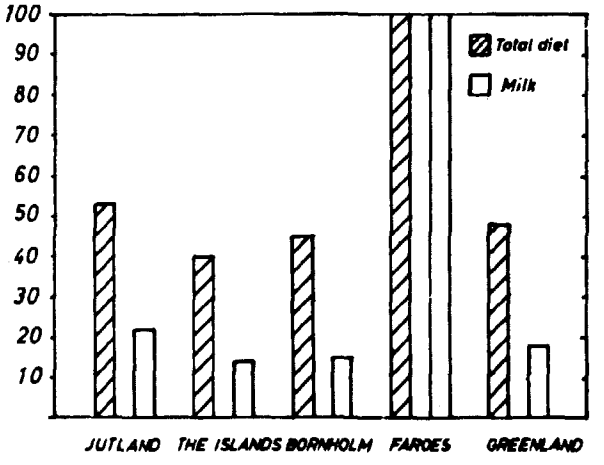

FIG. 4. The relative mean diet and milk ^{90}Sr/g Ca levels in 1962–1966 in Denmark, The Faroes and Greenland. (The Faroese levels = 100.)

and the low calcium content of the Faroese soil result in higher root-uptake in the crops and hence in higher ^{90}Sr concentrations in locally produced diet, e.g. in milk[15] than in Denmark, where the farming is intensive.

Infant bone levels estimated from tooth measurements

The newborn child contains about 30 g calcium and the 1-year-old infant 100 g.[21] Some remodeling, however, occurs and much of the original bone is thus reabsorbed.[22] Hence less than 30% of the bone in a one year old child is formed prenatally (and more than 70% *postnatally*).

A few measurements on infant bone from Western Eurpoe and U.S.A. collected in the period 1955–1958 have been performed by KULP *et al.*,[23] and by BAILEY *et al.*[22] As the Danish infant bone levels in 1963–65 have been rather similar to those in U.S.A. and Western Europe,[24] a comparison is made (cf. Table 5), where the mean levels in 1-year-old infant bone calculated from Refs. 22 and 23 are compared with the mean tooth levels in Denmark and Greenland.

If a correction for a possible external ^{90}Sr accretion is applied to the tooth figures in Table 5 by subtraction of 0.2 pCi ^{90}Sr/g Ca, the agreement with the measured bone levels becomes fairly good in 1955–1957. The discrepancy in 1958 might be due to the aforementioned incomplete tooth material from this year. The higher tooth than bone levels observed in 1955–1957 in Table 5 might, however, also be due to a relativcly higher ^{90}Sr level in Danish diet than in diets from Western Europe and U.S.A. in the period before 1958, the year when the aforementioned addition of $CaCO_3$ to Danish flour began.

The mean level of 1 year infant bone from Denmark was in 1964–1965 approx. 8 pCi ^{90}Sr/g Ca.[17] This has been the maximum level hitherto encountered in any group in human bone from Denmark. As the ^{90}Sr levels in deciduous teeth formed in 1950–1958 in Denmark, the Faroes and Greenland are proportional to the diet levels in 1962–1966 from these areas, and as the deciduous tooth levels approximately equal the 1-year-old infant bone levels, it is reasonable to assume (cf. Fig. 2), that the mean level in Faroese 1-year-old infant bone in 1964–1965 has been $4 \cdot 8 = 32$ pCi ^{90}Sr/g Ca. This level corresponds to an annual dose to the bone marrow in 1964–1965 of 22 mrad.[24]

The level in Greenland infant bone in 1964–1965 has probably been equal to the Danish level, hence the annual dose to the bone marrow in this case has been 6 mrad.

SUMMARY AND CONCLUSIONS

Since 1953 ^{90}Sr has been detectable in all types of shed deciduous teeth. From this year up until 1958 the level increased by a factor of four. The tooth level was nearly proportional to the milk level in the year when the child was born and in the subsequent year.

Second molars contained more ^{90}Sr than incisors and first molars. In accordance with

Table 5. A comparison of pCi ^{90}Sr/g Ca *levels in bone and teeth*

Year	1-yr-old infants bone in Western Europe and U.S.A.[22,23]			Shed deciduous teeth from Denmark and Greenland (from Table 1)		
	Mean	S.E.	Number of samples in the mean	Mean	S.E.	Number of samples in the mean
1953	—	—	—	0.5	0.04	26
1954	—	—	—	0.7	0.04	30
1955	0.7	0.2	8	1.0	0.04	26
1956	0.8	0.1	15	1.3	0.1	22
1957	1.3	0.1	21	1.5	0.1	18
1958	2.6	0.3	15	1.7	0.2	12

the longer postnatal calcification times of the former.

Faroese teeth contained four times more ^{90}Sr than teeth from Denmark and Greenland, the two latter did not differ significantly from each other. The variation between the different areas was proportional to the ^{90}Sr levels in a diet consisting of 75% milk and 25% total diet from these areas.

The ^{90}Sr mean level in shed deciduous teeth may provide an acceptable estimate of the ^{90}Sr bone level of the 1-year-old tooth donor.

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REFERENCES

1. L. C. REID, *Science* **134**, 1669 (1961).
2. H. L. ROSENTHAL, S. AUSTIN, S. O'NEILL, K. TAKEUCHI, J. T. BIRD and J. E. GILSTER, *Nature, Lond.* **203**, 615 (1964).
3. W. E. STARKEY, F. J. BRYANT and E. H. HENDERSON, *Int. Dent. Sourc.* **14**, 206 (1964).
4. M. MASSLER, I. SCHOUR and H. G. PONCHER, *Am. J. Dis. Child.* **62**, 33 (1941).
5. A. AARKROG and J. LIPPERT, Environmental Radioactivity in the Faroes 1962–66. Risö Reports Nos. 64, 86, 108, 131 and 155.
6. A. AARKROG and J. LIPPERT, Environmental Radioactivity in Greenland 1962–66. Risö Reports Nos. 65, 87, 109, 132 and 156.
7. F. S. BRYANT, A. MORGAN and G. S. SPICER, The Determination of Radiostrontium in Biological Materials, AERE-R-3030 (1959).
8. J. LIPPERT, Low Level Counting, Risö Report No. 44 (1963).
9. J. VESTERGAARD, Gier System Library No. 211, A/S Regnecentralen, Copenhagen (1964).
10. H. L. ROSENTHAL, J. E. GILSTER and J. T. BIRD, *Science* **140**, 176 (1963).
11. J. LINDEMANN, *Tandlægebladet* **69**, 297 (1965).
12. H. L. ROSENTHAL, J. T. BIRD, J. E. GILSTER, P. V. C. PINTO and S. O'NEILL, *J. Dent. Res.* **45**, 343 (1966).
13. HASL-142, p. 218 (1964).
14. J. H. HARLEY (editor), Manual of Standard Procedures, NYO-4700, Sec. Iss. 1967.
15. R. S. CAMBRAY, E. H. R. FISHER, W. L. BROOKS and D. H. PEIRSON, Radioactive fallout in Air and Rain, AERE-R-5260 (1966).
16. Meteorologisk Institut, Copenhagen, Ugeberetning om Nedbør m.m. (1953–58).
17. A. AARKROG and J. LIPPERT, Environmental Radioactivity in Denmark 1959–66, Risö Reports Nos. 14, 23, 41, 63, 85, 107, 130, and 154.
18. J. RIVERA and J. H. HARLEY, The HASL Bone Program 1961–64, HASL 163 (1965).
19. N. J. PETERSEN and L. D. SAMUELS, *Health Phys.* **12**, 1453 (1966).
20. J. LINDEMANN, *Tandlægebladet* **71**, 731 (1967).
21. H. H. MITCHELL, T. S. HAMILTON, P. R. STEGGERDA and H. W. BEAN, *J. Biol. Chem.* **158**, 625 (1945).
22. N. T. J. BAILEY, F. J. BRYANT and J. F. LOUTIT, Strontium 90 in Human Bone in the U.K. 1956–58. AERE-R-3299 (1960).
23. J. L. KULP and A. R. SCHULERT, *^{90}Sr in Man and His Environment II.* Columbia University, Palisades, New York (1961).
24. Report of United Nations Scientific Committee on the Effects of Atomic Radiation. New York (1966).

Health Physics Pergamon Press 1971. Vol. 21 (Dec.), pp. 803–809. Printed in Northern Ireland

PREDICTION MODELS FOR ^{90}Sr IN SHED DECIDUOUS TEETH AND INFANT BONE

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Abstract—Shed deciduous teeth were collected in 1966–69 in Denmark, the Faroes and Greenland from children born in the period 1953–63. 235 samples of crowns were analysed for ^{90}Sr. The ^{90}Sr levels in deciduous tooth crowns were related to the fall-out rate and the accumulated fall-out. The tooth levels in children born in 1950–62 could be described with the same equation as the ^{90}Sr bone levels in 1-yr-old infants born in 1962–68. The prediction models for ^{90}Sr in teeth and bones showed that for given amount of fall-out the Faroese levels became nearly twice as high as the Danish. The maximum teeth and bone levels were found in children born in 1963, where the Faroese level was estimated from the prediction model to be 24 pCi ^{90}Sr/g Ca.

INTRODUCTION

In an earlier paper[1] the results of ^{90}Sr measurements on shed deciduous teeth collected in 1962–65 in Denmark, the Faroes and Greenland were given. The present material was collected in the same areas in the period 1966–69 and is made up of teeth from children born until 1963. At that time the Danish infant bone measurements were initiated. Thus the gap between tooth and bone samples has been closed, at least for Denmark, and the calculation of prediction models more reliable than those used hitherto for infant bone and deciduous teeth has been possible.

MATERIALS AND METHODS

The samples were treated as previously.[1] However, the sizes of the samples analysed were smaller (2–5 g) this time as the activity levels were higher. Figure 1 shows that with approximation the ^{90}Sr levels were log normal distributed. It is evident that the Faroes belong to a distribution different from that of Denmark and Greenland. Hence the analysis of variance (anova) was carried out separately for the Faroes.

RESULTS AND DISCUSSION

Table 1 shows the decay-corrected pCi ^{90}Sr/g Ca levels in the samples of tooth crowns, and Tables 2 and 3 are the anovas of the natural logarithm of the figures.

Tooth crowns

The anova for Denmark and Greenland shows that the variation between areas was highly significant. The levels in eastern Denmark (the Danish islands and Bornholm) were 80% of the levels in Jutland and Greenland (cf. Fig. 2). This difference was not significant in the samples collected in 1962–65.[1] The observation is, however, in agreement with human bone measurements from Denmark,[2] where Jutland shows approx. 20% higher levels than eastern Denmark. It is not surprising that Greenland tooth levels are close to the Danish ones as the corresponding ^{90}Sr diet levels are very similar.[3,2] The Faroese tooth levels were, in accordance with earlier observations,[1] approx. 4 times as high as the Danish.

The variation between years was also highly significant. The levels from 1962 and 1963 show a steep increase as compared with the previous years. It is, however, interesting to notice that the increase was not so marked for the Faroes as for Denmark. This is a result of the greater dependence on accumulated fall-out in the Faroes (soil uptake) than in Denmark, where the diet levels follow the fall-out rate to a higher degree (direct contamination).[4]

Unlike the previous material[1] the present showed no significant difference between tooth types. However, incisors still showed the lowest levels. The anovas showed that no

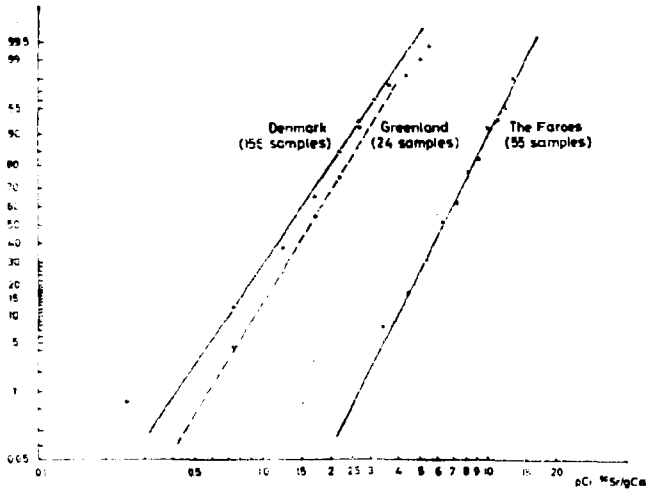

FIG. 1. The distribution of pCi ^{90}Sr/g Ca in shed deciduous tooth crowns from Denmark, Greenland and the Faroes (cf. Table 1).

interactions were significant, and the remainder was significantly lower than in the material from 1962–65 on account of the higher levels and the ensuing improved counting statistics.

As compared with the levels found in the teeth collected in 1962–65[1] the present samples were, for the same tooth type, area and year, not significantly different, although on the average the new material showed 10% higher levels.

Prediction models

In an earlier paper[5] prediction models for ^{90}Sr in human diet and tissue were calculated on the basis of data collected in Denmark (and the Faroes) in 1962–68. In the same paper a table for fall-out rate, d, in mCi $^{90}Sr/km^2/yr$ and accumulated fall-out, A, in mCi $^{90}Sr/km^2$ was given for the period 1950–68. On the basis of this table and on the measurements of infant bone collected since 1963[2] and on the present and earlier[1] tooth measurements, prediction models were calculated for ^{90}Sr in deciduous teeth and infant bone.*

The ^{90}Sr mean level in shed deciduous teeth has earlier[1] been considered as an estimate of the ^{90}Sr content in the tooth donor's bone when he was 1 yr old. If this model is adapted, we will consider bone from children 1–12 months old,† who died in the year $(i + 1)$, as representative of tooth donors born in the year (i), as regards their bone levels when they were 1 yr old. This assumption is justified because although the age group 1–12 months old has shown bone levels that are 10–15% lower than the 1 yr old group,[6] this is balanced out by the fact that the infant bone in this material came exclusively from Jutland where the levels are 10–15% higher than the country mean.[8] When the entire period since 1950 is considered, it is furthermore necessary to take the addition of creta praeparata to the Danish diet into account as this addition was first begun in April 1958. The creta praeparata raised the calcium level in the Danish diet by approximately 50%. Hence the pCi ^{90}Sr/g Ca tooth levels from children born until 1957 were recalculated with a multiplication by $\frac{2}{3}$. For 1957, which

* The tooth samples from children born in 1963 were not used in the calculations as the number of samples was too small.

† The yearly mean number of bone samples from the 1–12 month age group was 25 while it was only 3 for the 1–2 yr group; hence the former group was preferred.

Table 1. pCi ^{90}Sr/g Ca *in shed deciduous tooth crowns from Denmark, the Faroes and Greenland*

Year of birth of the cohort		1953	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963
	Incisors					1.53	2.23	2.09	1.74	1.80	5.32	5.99
						1.75	2.24	1.83	1.66	3.72		
			0.68	1.30	1.29	1.51	3.02	2.42	2.44			
	Cuspids			1.18	1.88	1.94	2.10	3.25	2.24			
						2.16	2.12	2.59				
						2.02						
Jutland	Molar		0.63	0.90	1.38	1.85	2.24	1.67	1.76	2.24		
	I		0.38	1.14	1.32	1.59		2.72	2.21			
					2.05	1.84		2.27				
		0.49	0.98	1.38	1.97	1.20	2.92	2.49	2.71	2.58		
	Molar				1.36	1.63	2.48	2.21	1.40			
	II				1.93	1.73	2.46	2.14				
					1.15							
	Incisors				1.87	1.89	2.52	1.40	1.67	1.43	2.46	4.04
							1.35	1.85	1.09	1.89		
			0.88	1.00	1.29	1.20	1.76	1.86	0.81	2.46		
	Cuspids			1.06	1.39		1.78	1.42	1.24	1.06		
					2.04		2.74					
							1.45					
The Danish islands				1.63	1.01	1.48	1.82	1.88	1.14	2.56		
	Molar			0.92	1.20	3.60	1.32	1.68	1.24			
	I				1.24		3.06	1.86				
							2.08					
		0.75	0.76	0.96	0.99	0.86	2.81	1.07	1.53	3.10	7.60	
	Molar		0.75	0.84	1.02	1.58	1.86	2.27	1.92	2.54		
	II				1.74		2.40	2.63				
							3.06					
	Incisors						2.12	2.00	1.19	1.51	3.38	
									1.10	1.64		
	Cuspids			1.79	1.54	1.66	2.30	2.70	1.05			
Bornholm	Molar			0.87	1.00	1.32	1.62	1.56				
	I						2.22					
	Molar											
	II			1.26	1.14	1.65	2.17					
	Incisors					6.17	10.90	5.80	5.24	6.36	8.25	
	Cuspids	3.59	3.36	5.82	5.53	6.64	6.98	8.30	5.12	8.03		
						7.67	6.80	11.35	10.30	8.97		
The Faroes	Molar	6.25	5.70	4.22	4.97	5.96	7.34	7.70	6.68	6.74	7.14	9.37
	I				4.99	12.50	8.59	10.33	7.93	14.30	10.90	
	Molar		3.12	3.79	4.90	9.75	5.82	8.56	6.96	9.53	11.00	13.52
	II				4.80	8.66	6.44	7.67	5.86	8.57	13.23	
	Incisors					2.70	1.74	3.22	3.04	1.94		
	Cuspids			1.22	1.55	2.13	2.22	2.88	1.91	2.96		
Greenland	Molar			0.83	1.28	1.91	1.77	2.51	2.18			
	I											
	Molar			1.47	1.67	1.44	2.26	2.30	1.43			
	II											

Table 2. Analysis of variance of the ln (pCi ^{90}Sr/g Ca) *in tooth crowns from Denmark (Jutland, and the Danish islands and Bornholm) and Greenland* (cf. Table 1)

Effect	Source	SSD	f	s^2	v^2	P
Main	Area	1.5578	3	0.5193	7.76	> 99.95%
	Year	20.0670	10	2.0067	30.00	> 99.95%
	Tooth type	0.3428	3	0.1143	1.71	—
2 factor interaction	Area × year	1.5864	23	0.0690	1.03	—
	Year × type	1.8571	20	0.0929	1.39	—
	Type × area	0.7014	9	0.0779	1.16	—
3 factor interaction	Area × year × type	1.9006	39	0.0487	0.73	—
Remainder		4.8148	72	0.0669		

Table 3. Analysis of variance of the ln (pCi ^{90}Sr/g Ca) *in tooth crowns from the Faroes* (cf. Table 1)

Effect	Source	SSD	f	s^2	v^2	P
Main	Year	4.1578	10	0.4158	7.42	> 99.95%
	Tooth type	0.2212	3	0.0737	1.32	—
2 factor interaction	Year × type	1.2356	22	0.0561	1.00	—
Remainder		1.0644	19	0.0560		

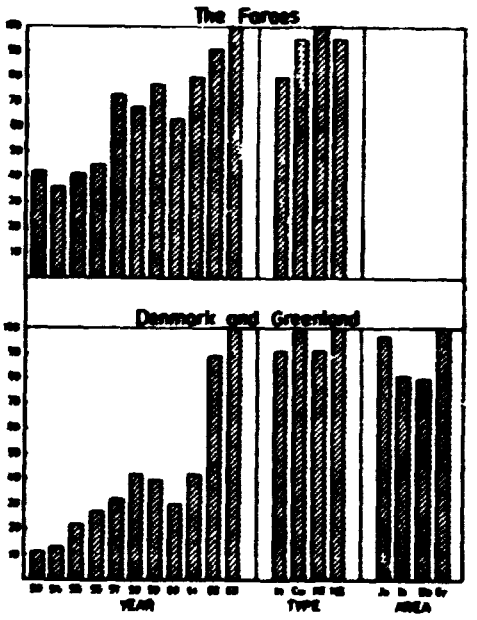

FIG. 2. Relative pCi ^{90}Sr/g Ca levels in shed deciduous tooth crowns from Denmark, Greenland and the Faroes. The Faroese levels were approximately 4 times as high as the Danish and the Greenland levels.

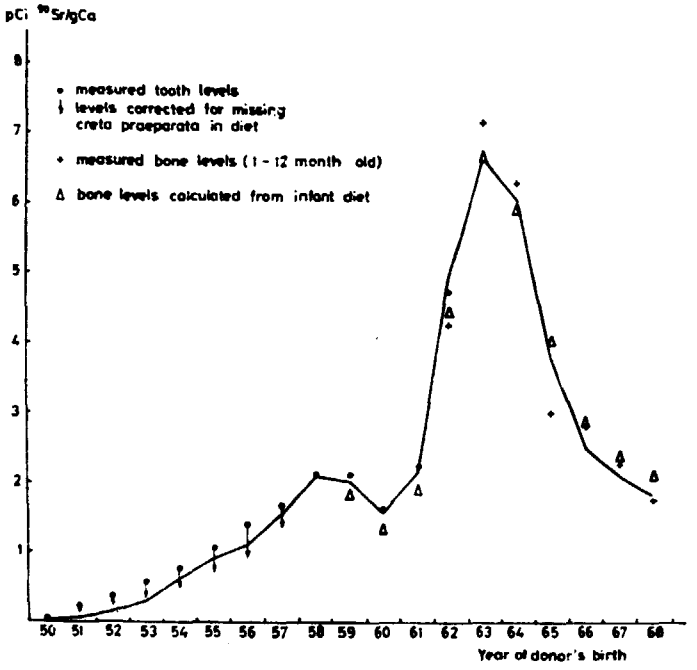

FIG. 3. Predicted (curve) and measured ^{90}Sr levels in deciduous tooth crowns and in infant (1 yr old) bone from Denmark. The multiple correlation coefficient, r, between observed and predicted levels was 0.9915, i.e. highly significant.

represents a transistional period, the multiplication factor was estimated to be $\frac{1}{3}$. The tooth levels used in the prediction models were the weighted sample means of the two sampling periods (1962–65 and 1966–69) corrected by the above-mentioned factors. For children born in 1962 the average between tooth crowns (4.69 pCi ^{90}Sr/g Ca) and 1–12 month old infant bone collected in 1963 (4.21 pCi ^{90}Sr/g Ca) was used.

The prediction equation for Denmark came out as:

$$(\text{pCi}^{90}\text{Sr/g Ca})_{\text{teeth or bone}} = 0.33d_{(i+i+1/2)} + 0.14d_{(i-1)} + 0.024A_{(i)},$$

where $d_{(i+i+1/2)}$ is the mean of the fall-out rates in the year of birth (i) and the following year, $d_{(i-1)}$ the fall-out rate in the year$_{(i-1)}$ and $A_{(i)}$ the accumulated fall-out by the end of the year of birth.

Figure 3 shows the prediction curve for Denmark compared with the measured (and creta-praeparata-corrected) tooth levels and with the measured bone levels.

It has earlier been assumed that the infants' diet during the first year of life corresponds to 75% milk and 25% total (adult) diet.[1] Let us suppose that the bone levels of the 1 yr old child (dead in the year $(i + 1)$) or of the tooth donor (born in the year (i)) are determined as the mean of the pCi ^{90}Sr/g Ca levels of the infant diet in the year (i) and $(i + 1)$. Let us further assume that the observed ratio between infant bone (or deciduous tooth crowns) and infant diet is 0.25, which is the normally accepted value for newly-formed bone in adults.[6] It is then possible to calculate the bone levels for the years 1959–68 if diet measurements are available. The calculated values from diet are also shown in Fig. 3. The multiple correlation coefficient, r, between the measured bone and tooth levels and the calculated levels from diet was 0.9731, i.e. highly significant. More refined models taking the pre- and

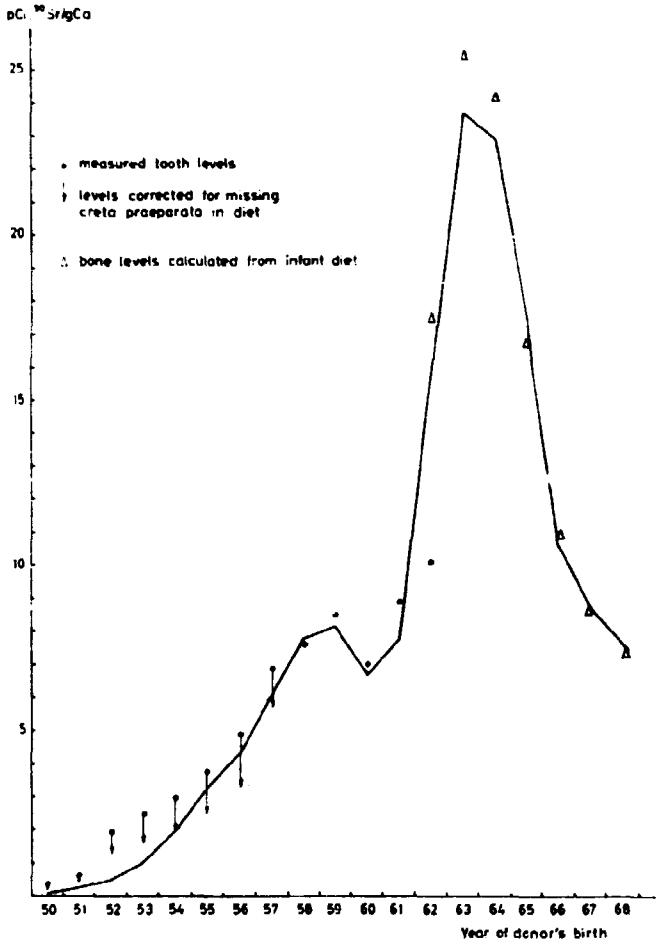

FIG. 4. Predicted (curve) and measured ^{90}Sr levels in deciduous tooth crowns and in infant (1 yr old) bone (calculated from infant diet, cf. the text) from the Faroes. The multiple correlation coefficient, r, between observed or calculated and predicted levels respectively was 0.9936, i.e. highly significant.

post-natal calcification and the varying discrimination factors for ^{90}Sr into account have earlier been applied.[1] The correlation shown in Fig. 3 indicates, however, that such refinements are hardly justified.

From the Faroes we have no measurements on infant bone. However, the bone levels could be calculated from the Faroese milk and total-diet determinations[4] in analogy with the above-mentioned procedure. From the measured ^{90}Sr levels in deciduous teeth crowns (recalculated with respect to missing creta praeparata in the diet as described above for Denmark) and from the calculated bone levels the prediction equation:

$$(\text{pCi } ^{90}\text{Sr/g Ca})_{\text{teeth or bone}} = 0.48d_{(i+i+1/2)} + 0.33d_{(i-1)} + 0.046A_{(i)}$$

shown in Fig. 4 was obtained. The fall-out data were as for Denmark obtained from Ref. 5.

A comparison between the Danish and Faroese prediction models shows that for a given amount of fall-out the Faroese bone levels become nearly twice as high as the Danish. Similar observations have earlier been made for milk from the two areas,[5] where the Faroese ^{90}Sr levels for a given amount of fall-out were nearly 3 times as high as the Danish levels. The difference between milk from the two areas is greater than the bone difference because beside Faroese-produced milk the infant diet in the Faroes contains milk as well as other diet components imported from Denmark.

In the first report on deciduous teeth[1] the Faroese infant bone levels in 1964–65 were estimated at 32 pCi ^{90}Sr/g Ca. This estimate was based on the assumption that the Faroese levels were 4 times as high as the Danish. During periods with high fall-out rates as in 1963–64 this assumption does, however, not come true. The prediction equations in Figs. 3 and 4 show that the ratio between the Faroese and Danish rate factors is less than the ratios for the "lag" and the cumulative factors. Hence, during periods with intensive fall-out where the rate term in the equations plays an important role for the ^{90}Sr levels, the ratio between Faroese and Danish bone levels becomes less than 4 (cf. also Fig. 2).

CONCLUSION

The agreement between predicted and observed tooth and bone levels (cf. Figs 3 and 4) supports the theory that the pCi ^{90}Sr/g Ca ratio in deciduous tooth crowns equals the bone level of 1-yr old infants and vice versa. The agreement furthermore indicates that the earlier estimated fall-out levels for the period 1950–61[6] have not been far off the actual fall-out situation in Denmark and the Faroes in this period Furthermore the use of an infant diet model consisting of 75% milk and 25% adult diet for the calculation of the tooth and bone levels on the basis of an observed ratio between bone tissue and diet of C.25 has been shown to be justified.

Acknowledgement—The study is still based on the co-operation of teachers and pupils of the 25 schools in Denmark the Faroes and Greenland, which participate in this programme, to all of whom thanks are due.

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REFERENCES

1. A. AARKROG, *Health Phys.* **15**, 105 (1968).
2. Risö Reports Nos. 23, 41, 63, 85, 107, 130, 154, 180, 201 and 220 (1969–70).
3. Risö Reports Nos. 65, 87, 109, 132, 156, 182, 203 and 222 (1963–70).
4. Risö Reports Nos. 64, 86, 108, 131, 155, 181, 202 and 221 (1963–70).
5. A. AARKROG, *Health Phys.* **20**, 297 (1971).
6. Reports of UNSCEAR. New York (1962, 1964, 1966 and 1969).

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CÆSIUM-137 FROM FALL-OUT IN HUMAN MILK

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IT is well known that in human milk the ratio between strontium-90 and calcium is lower than in the diet of the mother[1]. Only sparse information exists as regards a comperison of the contents of cæsium-137 (the other long-lived biologically significant fission product) in a mother's milk and in her diet. Several animal experiments have been performed by Wasserman, Comar and Twardock[2] and have shown that the $^{137}Cs/K$ ratio in goats' milk is 1·4 based on a dietary $^{137}Cs/K$ ratio of unity.

The following measurements were performed with milk from a primipara 24-year-old Danish woman, who bore a child October 15, 1961. The milk was collected daily during the period November 1, 1961–September 10, 1962. Until the last half of August 1962 the daily mean production of milk was about 1½ l.; of this, about ¾ l. per day was consumed by her baby; the rest was obtained for analyses. The milk used for analyses was identical to the milk used for the feeding of the baby, as the mother drew out all milk before each feeding and fed the baby from a feeding-bottle. Most of the daily samples were dried and ashed (at 400°–450° C), and the ash was bulked into monthly samples before measurement. These ashed samples have been measured in a 3 in. × 4 in. well-type sodium iodide (Tl) crystal (Harshaw) with a 50-ml. well. Several samples were, however, measured as fresh milk in a 1·6 l. can, which fits around a 3 in. × 3 in. sodium iodide (Tl) crystal (Harshaw). The pulses were analysed in a *TMC256* channel analyser.

Table 1 shows the results of the monthly measurements and gives furthermore, for comparison, the levels of monthly collected dried milk samples from dairy cows living in the same part of the country as the woman.

Table 1 shows that the human milk (as well as the cows' milk) displays a variation in content of cæsium-137 with time. In the winter and in early spring, before the dairy cows started grazing, the human milk-levels were rather constant: 8·4 ± 0·5 pc. $^{137}Cs/g$ K. In May the level rose by a factor of nearly four, and in the following three months, June–August, the level was nearly constant: 34 pc. $^{137}Cs/g$ K. Already in September, Danish dairy cows start to have an increasing amount of feed other than grass, and the effect of this decreasing contribution from grazing was observed both in the human milk and in the cows' milk collected in September.

Table 1. Caesium-137 and Potassium in Milk

Sampling period	Human milk pc. ^{137}Cs/g K	Human milk g K/l milk	Cows' milk pc. ^{137}Cs/g K
November 1–December 31, 1961	8·1	0·23	4·5
January 1–31, 1962	9·2	0·21	1·5
February 1–28, 1962	7·1	0·28	2·0
March 1–31, 1962	7·4	0·25	3·5
April 1–26, 1962	10·5	0·28	4·0
May 23–31, 1962	29·6	0·24	22·0
June 1–30, 1962	34·0	0·27	33·0
July 1–31, 1962	34·0	0·28	36·0
August 1–31, 1962	33·7	0·29	37·0
September 1–10, 1962	28·1	0·31	29·0

The relative S.D. due to the measurements of the human milk is less than 10 per cent.

As the content of caesium-137 in the human milk appeared to be nearly constant in the period November–March (Table 1), and as the mother was on a regular diet in this period (1 kg dairy products, 0·15 kg grain products, 0·3 kg fruit and vegetables, 0·2 kg meat, fish and eggs, and 0·6 kg water as tea or coffee), it was anticipated that a steady state between the mother, her diet and her milk had been reached. The mother's diet and her milk were gathered for one week in March. The diet-sampling was performed by having the mother prepare double portions of everything she consumed in the week the experiment ran. The duplicates were put into a pail, which was sent to the laboratory. Six months later, in the beginning of September, the experiment was repeated, but unfortunately the milk-production was decreasing rapidly in this period, and after September 10 only very little milk was secreted. Table 2 shows the results from the two periods.

Table 2 shows that the ^{137}Cs/K ratio is higher in the donor's milk than in her diet. The ratios between caesium-137 secreted by the milk and ingested with the diet are related to the amount of milk secreted. Thus the percentages of ingested caesium-137 secreted per litre milk only differ slightly for the two experimental periods, March and September. As probable explanations for the relatively large differences between the observed ratios between milk and diet from the two months, beside the differences due to variation in milk production, one could mention that the daily average intake of potassium with diet in March was 2·8 g against 2·2 g in September, and that the concentration of potassium in the human milk from September was about 20 per cent higher than in the milk from March; but most important, that the September period was just before the end of the lactation period and thus could not be assumed as representing a steady state.

To get an estimate of how much and how long a time it would take for a single oral caesium-137 dose given by the food to be excreted in the milk, 260 g of reindeer meat from Greenland was given to the milk donor on April 26. Lidén[3] has shown that reindeer meat has a relatively high caesium-137 content, and thus is suited as a tracer in human biological experiments. The total content of caesium-137 in the 260 g of meat was measured at 8,200 ±

Table 2. A Comparison between the Levels of Cæsium-137 in Human Milk and in the Diet of the Donor

Sampling period	Sample	pc. ^{137}Cs/g K	pc. ^{137}Cs secreted daily in the milk	pc. ^{137}Cs ingested daily with diet	$\frac{\text{(pc. }^{137}\text{Cs/g K) milk}}{\text{(pc. }^{137}\text{Cs/g K) diet}}$	$\frac{\text{pc. }^{137}\text{Cs secreted}}{\text{pc. }^{137}\text{Cs ingested}}$	Ingested ^{137}Cs secreted per l. milk (per cent)
March 10–16, 1962	Milk	9·2 ± 0·2	6 ± 1	—	1·9 ± 0·1	0·43 ± 0·08	32 ± 6
March 9–15, 1962	Diet	4·9 ± 0·2	—	14 ± 1			
September 4–10, 1962	Milk	27·4 ± 1·5	9 ± 2	—	1·1 ± 0·1	0·17 ± 0·04	28 ± 7
September 3–11, 1962	Diet	24·2 ± 0·5	—	53 ± 5			

The error terms in the table indicate one *S.D.* due to the measurement.

Table 3. CAESIUM-137 LEVELS IN HUMAN MILK AFTER A SINGLE ORAL INTAKE OF 8,300 pc. CAESIUM-137

Sampling period	pc. ^{137}Cs/g K	pc. ^{137}Cs/l.	Estimated ^{137}Cs content in the milk from the fall-out background: pc. ^{137}Cs/l.	Accumulated excretion in the milk corrected for fall-out background: pc. ^{137}Cs	Accumulated excretion percentage
April 27–29	220	97		372	4·5
April 30–May 2	93	42	4	524	6·3
May 3–5	80	33		640	7·7
May 6–8	65	29		732	8·8
May 9–11	55	25	5	812	9·8
May 12–14	44	20		872	10·5
May 15–17	48	23	14	922	11·1
May 18–22	30	14		922	11·1
May 23–31	31	15	15	—	—
June	34	16	16	—	—

The relative *S.D.* of the results due to measurements is less than 10 per cent.

55 pc. The milk was collected and measured (mostly as fresh milk) during the month following intake. Table 3 shows the results.

This experiment was complicated by the fact that the dairy cows in the part of the country where the milk donor is living began grazing about the middle of May this year and consequently the 'normal' caesium-137 content in the mother's milk increased by a factor of nearly 4 (Table 1) during the experiment. However, it is evident from the figures in Table 3 that about 7 per cent of the ingested dose of caesium-137 was excreted in the milk within the first week after the intake of the reindeer meat, and within a fortnight about 10 per cent was excreted by the milk. For dairy cows, it has been shown[4] that about 10 per cent of a single oral dose of radiocaesium will, in a 7 days collection period, be recovered in the milk.

The present investigation indicates that babies fed on human milk could be exposed to a greater caesium-137 body burden than if they were fed on milk from dairy cows, provided that the diet of the mother shows a greater (about 2–3 times) ^{137}Cs/K ratio than the cows' milk. It should be mentioned that the specific caesium-137 activity per litre cows' milk normally will be greater than in human milk, even if the ^{137}Cs/K ratio is greater in the human milk. This is due to the fact that the concentration of potassium in human milk is only about one-third the concentration of potassium in cows' milk.

[1] Lough, S. A., Hamada, G. H., and Comar, C. L., *Proc. Soc. Exp. Biol. Med.*, **106**, 196 (1960).

[2] Wasserman, R. H., Comar, C. L., and Twardock, A. R., *Intern. J. Rad. Biol.*, **4**, 299 (1961).

[3] Libbin, K., *Acta Radiologica*, **56**, 237 (1961), and personal communication (1962).

[4] Langemann, F. W., and Comar, C. L., presented at FAO/WHO/IAEA seminar, December 11–15, 1961.

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